

*THE INFLUENCE OF NATURAL
CONVECTION AND CHANGES IN
THE PHYSICAL PROPERTIES ON
THE LIMITS OF FLAMMABILITY*

GÖRAN S HOLMSTEDT

DEPARTMENT OF PHYSICS

LUND INSTITUTE OF TECHNOLOGY

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THE LIMITS OF FLAMMABILITY

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ABSTRACT

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FUND INSTITUTE OF TECHNOLOGY

ABSTRACT

Thesis LUTF D2 (TF AF 1002)/ 1- 85 (1979)

Title: The influence of natural convection and changes in the physical properties on the limits of flammability.

Autor: Göran S Holmstedt

Department of Physics, Lund Institute
of Technology, S 220 07 Lund 7, Sweden.

Abstract. This thesis, based on four papers, deals with the behaviour of flames in pre-mixed gases near the limits of flammability. In paper I, the effect of a change in the initial pressure and in the amount of an inert gas added on the limits of flammability in a cylindrical vessel is studied. This report also contains a comparison between different ignition sources. Hot and fusing wires were found to be more efficient than electric sparks in igniting near limit mixtures. In paper II, an ignition source which is capable of heating fine wires (0.02-0.2 mm in diam) to temperatures up to 2000K within a millisecond and keeping the temperature within fixed limits for periods of seconds is described.

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The source is easy to calibrate and can also be used as a detector of ignition delay times. In paper III, the temperature field in a fluid surrounding a horizontal wire which is suddenly heated has been studied with a Mach-Zehnder interferometer and a high-speed photographic recording system. It has been found experimentally that the outer isotherms start to move upwards immediately with a constant acceleration for about 100 msec. A simple model predicts the acceleration as a function of the energy dissipation in the wire and the transport properties in the fluid. In paper IV, this ignition source is used in an investigation of flame behaviour near the flammability limits. The growth of an ignition kernel is studied perpendicular to the wire with a Schlieren system and along the wire with a Mach-Zehnder interferometer. The influence of convection currents is considered by measuring ignition delay times and laminar burning velocities near the flammability limits. By accelerating the wire, the convection currents during the pre-ignition period can be delayed.

Key words: Limits of flammability, Ignition, Burning velocity, Temperature measurements, Natural convection, H_2-O_2 , H_2 -air.

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Signature *Göran Holmstedt* Date *1/5 1979*

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 <i>PAPER II</i>	 A line heat source and detector for transient convection and ignition experiments. G.Holmstedt Submitted to Physica Scripta
 <i>PAPER III</i>	 Experimental investigation of the onset of free convection near a suddenly heated horizontal wire. G.Holmstedt Submitted to Int.J.Heat Mass Transfer
 <i>PAPER IV</i>	 The ignition of reactive gases near the flammability limits by heated wires.

INTRODUCTION

Premixed combustible gases are known to lose their capacity for free flame propagation on depletion or enrichment of the combustible content of the gas mixture. An external ignition source has to be supplied as the mixtures are incapable of spontaneous ignition. Ideally, these limits of flammability are independent of the size and power of the ignition source. In reality, the boundary is not well defined and a transition range sometimes of variable width is observed in experiments. Flammability limits have been studied experimentally mainly by using propagating flames. Comprehensive reviews are given by Coward and Jones [1] and Lovachev et al. [2]. Experimental studies with freely propagating flames of the part played by convection have revealed a large number of flammability problems. Some investigators consider convection to promote flame propagation [3] and others that it inhibits it [4,5]. Recently, Yamaoka and Tsuji [6] have stated that the limits of flammability determined by using a counterflow flame only depend on the physicochemical properties of the mixture. Many investigators believe in the existence of fundamental limits, while others consider they are due to the experimental conditions [7]

At the same time, flammability limits are needed for preventing explosion hazards. Many theories about the limits of flammability have been proposed [8,9,10,11], the only one including convection being due to Lovachev [12]. An alternative way is to use thermal ignition theory applied to gases [13] or to compute time-dependent laminar flame structure [14]. Common to these methods is that they cannot at present predict experimental values.

The experiments described in this thesis have been performed to study the influence of natural convection and changes in physical properties on the flammability limits. Particular attention has been paid to the ignition source, to nonstationary phenomena in the formation of the flame front and to how the convective motion of the hot gases develops in time. In most of the experiments, hydrogen has been used for several reasons:

- a) The hydrogen-oxygen reaction has often been used to test different theoretical flame models and many data are known.
- b) Hydrogen is considered to be an alternative to the hydrocarbons as an energy carrier in the future with increased demands for its safe handling.
- c) Hydrogen mixed with 2-3 vol o/o of oxygen may be used as a breathing gas for deep sea diving. Experiments with animals are at present in progress at the Department of Physiology, University of Lund.

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PAPER 1

The Upper Limit of Flammability of Hydrogen in Air, Oxygen, and Oxygen-Inert Mixtures at Elevated Pressures

Göran S. Holmstedt

Department of Physics, Lund Institute of Technology, Lund 7, Sweden

The upper limit of flammability of hydrogen in air, oxygen, oxygen-helium, oxygen-neon, oxygen-argon and oxygen-carbon dioxide mixtures was measured at room temperature and pressures between 0.97 and 29 atm in two cylindrical bombs with volumes of 1.5 and 5.2 liters. The limit in ternary mixtures was determined in 20%, 40%, 60% and 80% helium, 20% neon and argon and 10% carbon dioxide concentrations. The maximum safe percentage of oxygen in a hydrogen-oxygen-helium mixture was calculated for pressures between 0.97 and 29 atm.

Introduction

In recent years, deep-sea divers have used helium-oxygen mixtures as breathing gas in attempts to reach greater depths in sea diving. As even helium-oxygen mixtures become too dense for comfortable breathing at depths greater than 200 meters of sea-water, it has been suggested that the helium should partially or entirely be replaced by hydrogen, since the breathing work is proportional to the square root of the density of the gas. Limited tests carried out by Arne Zetterström [1] and Ralph W. Bauer [2] indicate that hydrogen may not be toxic at depths to about 500 meters of sea water. To ensure the safe operation of hydrogen-oxygen mixtures, it is necessary to know the influence of pressure, temperature, and inert gases on the upper limit of flammability.

Most of the papers about the hydrogen-oxygen reaction and the limits of flammability deal with experiments performed at atmospheric and sub-atmospheric pressures. The few values available on the upper limit of flammability at elevated pressures differ greatly, depending both on the experimental apparatus

used and the definition of the limit of flammability. The present experiments have been carried out to determine the influence of pressure on the upper limit of flammability of hydrogen-air, hydrogen-oxygen, and hydrogen-oxygen-inert mixtures at room temperature. The inert gases used have been helium, neon, argon, and carbon dioxide. From the results, the maximum safe percentage of oxygen in a hydrogen-oxygen-helium mixture at room temperature has been calculated for pressures between 0.97 and 29 atm. The upper limit of flammability is given as the minimum oxygen percentage by volume in this paper, contrary to the usual maximum fuel percentage, and the pressure as atmospheres abs.

Experimental Method

The apparatus used is shown diagrammatically in Fig. 1. The gas mixtures were tested for flammability in a cylindrical cavity situated in the center of a 10.5-cm diameter gun barrel, which had been reduced in length and plugged with a cone of brass. The diameter of the cavity was 12 cm, and the volume could be chosen as

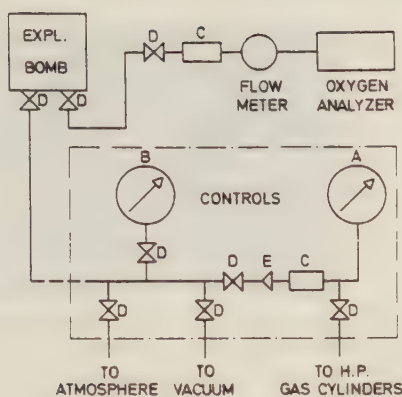


Figure 1. High pressure explosion apparatus. A, pressure gauge; B, precision pressure gauges; C, driers; D, valves; E, non-return valve.

either 1.5 or 5.2 liters. The bomb was designed to withstand explosion pressures of 1900 atm, the tubes 580 atm, and the valves 430 atm. The bomb, the gas supply, and the controls were situated in different rooms. After being dried, the gas mixtures were prepared in the bomb by the method of partial pressures using a mercury manometer with an accuracy of 0.2% for pressures up to 3 atm, and a set of Nordarmatur precision gauges with an accuracy of 0.5% at higher pressures. The gases were added to the bomb in the reverse order of their concentrations in the final mixture. Mixing in the bomb was achieved by means of a mechanical agitator consisting of a disk of brass running on two bars of brass which were parted 9 cm. By using the gun's elevation system, the agitator was allowed to move 40 times through the mixture during the 90–120 min which passed before the mixtures were tested for flammability. The compositions of the mixtures were calculated directly from the partial pressures, and compressibility effects were neglected. The mixing procedure, checked with a Beckman E2 oxygen analyzer which measured the magnetic susceptibility of the mixture with a magnetic torsion gauge, was such that the concentrations of the gases were accurate to ± 0.1 vol% for the oxygen and ± 0.5 vol% for the inert gases and the hydrogen in ternary mixtures.

Ignition

The gas mixtures were tested for flammability with electric sparks and fusing wires. Electric sparks were supplied to the gas mixtures across two 1-mm hemispherical electrodes of aluminium with an electrode distance of 1.3 mm. The electrode holders were situated axially 5 cm above one end of the bomb. The electric circuit consisted of a set of condensers with a total electrostatic capacity of $0.012 \mu\text{F}$ which was charged by a high voltage dc supply through a resistance of $30 \text{ M}\Omega$ and discharged over the electrodes either directly or with a series resistance of 250, 2000, or $10,000 \Omega$ in order to extend the discharge time. Condensers and conductors were designed to withstand 25 kV. From determination of the breakdown voltage at different pressures the input energies have been calculated and are given in Table 1.

If combustion did not occur after four successive sparks, two wires 5 cm in length were fused simultaneously. One of the wires was made of nickelin of 0.05 mm diameter and the other of kanthal of 0.10 mm diameter. Both wires were mounted on a holder situated axially 3 cm above one end of the bomb and bowed in a single loop against the center. The wires were fused when they were connected in series with a resistance to an ac transformer. The voltage fall across the resistance in series was measured with a cathode ray oscilloscope and the trace was photographed. In this way current intensities and input energies could be determined since changes of the transformer voltage and the resistance in series were negligible. In all experiments the transformer voltage was

Table 1. Breakdown Voltages and Input Energies ($\frac{1}{2} \text{ CV}^2$) at Elevated Pressures

Mixture composition: 75 vol% hydrogen, 20 vol% argon, and 5 vol% oxygen. Electrode distance, 1.3 mm.

Pressure (atm)	1.94	2.90	4.84	9.7	19.4	29.0
Breakdown voltage (kV)	5	6	9	14.5	19	25
Input energy (J)	0.15	0.2	0.7	1.2	2.1	3.7

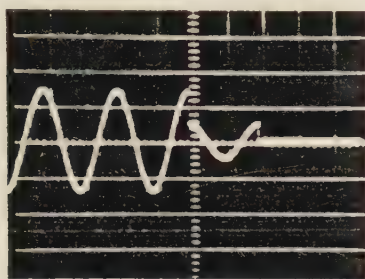


Figure 2. A trace of the current intensity when two wires are fused at an initial pressure of 29 atm. Mixture composition: 80 vol% helium, 15.5 vol% hydrogen and 4.5 vol% oxygen. One square corresponds to 5 A and 10 msec.

34 V and, in consequence, an energy of 5–8 J was released in the mixture during 60–90 msec. A typical trace is shown in Fig. 2.

Combustion Criteria

Measurements of the temperature and pressure were used to indicate the occurrence of flammability. Temperatures were measured with a thin Pt/Pt, 10% Rh thermocouple inserted in the bomb, and pressure falls were measured with precision pressure gauges when the gas mixtures had cooled down. Sometimes the oxygen concentration of the residual gas was checked with the paramagnetic analyzer.

The limit of flammability was determined by the method of trial and error. It was found that the transition from complete combustion to no reaction occurred at a decrease of the oxygen concentration by 0.1 vol% in most of the determinations of the upper limit. In these cases the upper limit was taken as the highest oxygen concentration in the mixtures which did not react. In the few remaining determinations at which a decrease of the oxygen concentration by 0.1 vol% gave rise to a transition from complete to incomplete combustion, and another decrease by 0.1 vol% resulted in a transition to no reaction, the upper limit was taken as the oxygen concentration for incomplete combustion.

Results

The flammability tests were carried out with dried gas mixtures at an initial temperature of

$21.5 \pm 1.5^\circ\text{C}$. It was found that the fusing wires were more efficient in igniting oxygen-lean mixtures than the electric sparks. Flammability tests with only one fusing wire indicated that nickelin and kanthal wires had the same efficiency in the ignition of hydrogen-oxygen mixtures at 4.84 atm pressure. The remaining tests were carried out with simultaneous fusing of two wires. Flammability tests with hydrogen-air, hydrogen-oxygen and hydrogen-oxygen-helium mixtures of 20% helium concentration at 0.97, 2.90, and 4.84 atm pressure gave identical upper limits in two bombs of 1.5 and 5.2 liters in volume. The remaining tests of a total of 187 measurements were carried out exclusively in the 1.5 liter bomb.

The work has been divided into three sections: the first contains hydrogen-oxygen and hydrogen-oxygen-helium, the second hydrogen-oxygen-inert of 20% inert concentration, and the third hydrogen-air and hydrogen-oxygen-carbon dioxide mixtures.

Flammability of Hydrogen in Oxygen and Oxygen-Helium Mixtures

The experiments were carried out with hydrogen-oxygen-helium mixtures of 0, 20, 40, 60 and 80% helium concentration at 0.97, 1.94, 2.90, 4.84, 9.7, 19.4, and 29.0 atm pressure. The influence of the pressure and the diluent on the upper limit of flammability is shown in Fig. 3. By determining the point of intersection between the tangents to the flammability curves in a diagram where the limits had been plotted against the dilution, the critical oxygen concentration was given on a fuel-free basis. The critical oxygen concentration represents the maximum percentage of oxygen which will be safe in any unknown mixture of hydrogen with oxygen and helium at room temperature and is given by the dashed line in Fig. 3. The lower limit of flammability which usually varies a little with the dilution was at this determination assumed to be constant, being 5 vol% hydrogen at all pressures and helium concentrations. The critical oxygen concentration,

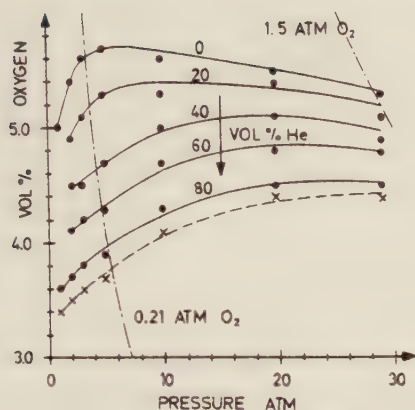


Figure 3. The upper limit of flammability of hydrogen-oxygen-helium mixtures of 0, 20, 40, 60, and 80% helium concentration at elevated pressures. -----, maximum safe percentage of oxygen; - - - - - isobars showing partial pressures of oxygen of 0.21 and 1.5 atm.

however, would only have been affected by ± 0.1 vol% if any other lower limit of flammability between 2 and 10 vol% hydrogen had been chosen. To check the maximum safe oxygen concentration, some mixtures with compositions near the intersection point at 1.94 atm pressure were tested for flammability. No reaction occurred for hydrogen in 90 vol% helium with the vol% of oxygen varying from 3.4 to 5.2 in steps of 0.2%. The dashed and dotted lines in Fig. 3 represent the limits of acceptable partial pressures of oxygen for breathing.

Out of the papers about the upper limit of flammability of hydrogen-oxygen mixtures at room temperature, a few give a lower oxygen concentration than the values given in Fig. 3. Van Diest and De Greef [3] give an upper limit of 4 vol% oxygen at atmospheric pressure, and Yantovskii and Chernyak [4] give a constant upper limit of 4.5 vol% oxygen and a zone of incomplete combustion between 7 and 4.5 vol% oxygen for pressures between 1 and 20 atm. Since the transition from complete combustion to no reaction occurred directly in these experiments in agreement with Refs. 2, 5, 6 and 7, the discrepancy is probably due to different choices of experimental conditions.

Table 2. The Lowest Oxygen Concentration of the Hydrogen-Oxygen Mixtures of Dorr and Schreiner [2]^a

Initial Pressure (atm)	1	4	7	16	34	42
Lowest vol% of oxygen	5.6	5.8	6.0	6.0	5.4	6.1

^a Inflamed mixtures at those pressures where the results of these authors are given in most detail.

Recently Dorr and Schreiner [2] have carried out flammability tests with hydrogen-oxygen mixtures at pressures between 1 and 42 atm. Their results are given in Table 2.

Flammability of Hydrogen in Oxygen-20 Vol% Inert Mixtures

In Table 3 the effect of the pressure on the upper limit of flammability of hydrogen in oxygen-inert mixtures of 20% inert concentration is shown. At pressures above 5 atm the different inert gases gave the same limit within the experimental accuracy. In Fig. 4 the values from Egerton and Powling [5] are included for comparison.

Flammability of Hydrogen in Air and Hydrogen in Oxygen-10 Vol% Carbon Dioxide Mixtures

In Fig. 5 the effect of the pressure on the upper limit of flammability of hydrogen in air and hydrogen in oxygen-carbon dioxide mixtures of 10 per cent carbon dioxide concentration is shown. The oxygen concentration at the upper limit of flammability of hydrogen-air mixtures given in Fig. 5 over the whole pressure range are lower than the values shown for small closed bombs in Refs. 6-9. At atmospheric pressure

Table 3. Volume % of Oxygen at the Upper Limit of Flammability of Hydrogen in Oxygen-20 Vol% Inert Mixtures at Elevated Pressures

Pressure (atm)	1.94	2.90	4.84	9.7	19.4	29.0
Helium	4.9	5.1	5.3	5.3	5.4	5.1
Inert	Neon	4.9	5.2	5.4	...	...
	Argon	4.7	4.8	5.1	5.4	5.4

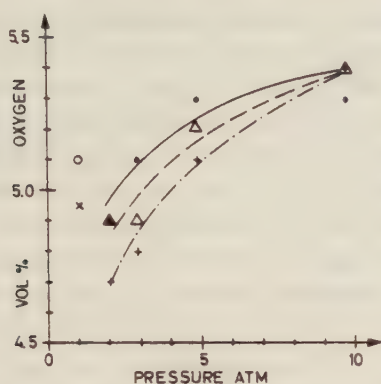


Figure 4. The upper limit of flammability of hydrogen-oxygen-inert mixtures of 20% inert concentration at elevated pressures. — (●) helium; --- (Δ) neon; - · - · - (+) argon, this work. ○, 19 vol% helium at 1 atm pressure Egerton and Powling [5]; ×, 18 vol% argon at 1 atm pressure Egerton and Powling [5].

the limit is in tolerably good agreement with the values obtained in an open tube by Egerton and Powling [5] and in a closed 1-liter bomb by Eitner [8].

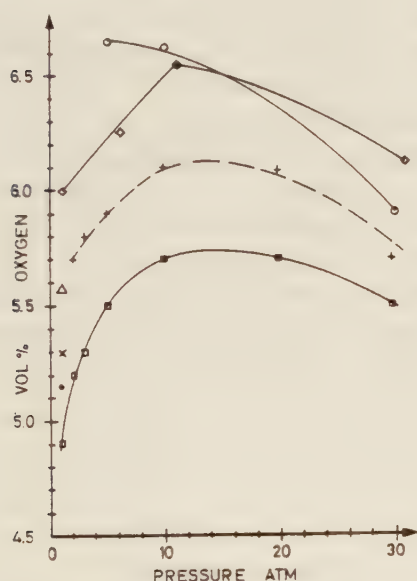


Figure 5. The upper limit of flammability of hydrogen-air and hydrogen-oxygen-10 vol% carbon dioxide mixtures at elevated pressures: □, Hydrogen-air, this work; ×, hydrogen-air, Egerton and Powling [5]; ○, hydrogen-air, Bone, Newitt and Smith [6]; ◇, hydrogen-air, Berl and Werner [7]; ●, hydrogen-air, Eitner [8]; Δ, hydrogen-air, Weissweiler [9]; +, Hydrogen-oxygen-10 vol% carbon dioxide—this work.

Discussion

Theories of flame propagation which include heat-transfer and diffusion processes lead to conservation equations which can in principle be solved to give a burning velocity if physical and chemical data are known. These equations do not, however, explain discontinuous phenomena such as limits of flammability. Weinberg [10] has used an alternative approach, suggesting that the reaction system is rate-controlled by a single chain-branching and a single chain-breaking step. This approach has been applied to the hydrogen-oxygen reaction by Butlin and Simmons [11], who have studied the effect of hydrogen halides on the limits of flammability. The approach seems to explain the effect of iodide and bromide, which act as chemical inhibitors, but does not make predictions for the uninhibited case. Since flammability limits cannot be predicted with certainty, one has recourse to experiments in which one aims at determining the limits in an apparatus-independent manner.

Coward and Jones [8] have discussed the choice of experimental conditions in their critical review of limits of flammability. They recommend a combustion tube diameter of at least 5 cm for most flammable mixtures. If the tube is closed, the length may sometimes affect the result. Terrada et al. [12], however, found that the length did not affect the extent to which the hydrogen-oxygen reaction proceeded in a closed tube of 1 cm diameter for mixtures near the upper limit of flammability. Identical upper limits were obtained in these experiments in two tubes of 12 cm in diameter and 13 cm and 46 cm in length. The sources of ignition which have been used most frequently in flammability tests are electric sparks and small flames at atmospheric pressure, and hot and fusing wires at elevated pressures [6, 7, 13, 14]. Lewis and von Elbe [15] and Rose and Priede [16], among others, have studied spark ignition of hydrogen-air mixtures and determined the minimum ignition energy and the quenching distance at atmospheric pressure as a func-

tion of the geometry and the material of the electrodes, the mixture composition, and the electric circuit. The quenching distance is several millimeters long, and the minimum ignition energy is several millijoules for hydrogen-air mixtures near the upper limit of flammability at atmospheric pressure. The fact that fusing wires were more efficient in igniting oxygen-lean mixtures in these experiments indicates that the quenching distance for near-limit hydrogen-oxygen-inert mixtures is longer than 1.3 mm, even at pressures up to 29 atm, since the energies of the sparks were most likely larger than the minimum ignition energy.

Stout and Jones [17] have studied the ignition of a hydrogen-air mixture by fusing wires of different diameters but their extrapolated value of the minimum ignition energy is in poor agreement with values from direct determinations with electric sparks, as has been pointed out in Ref. 15. The wires made of kanthal and nickelin used in these experiments had the same ignition efficiency at 4.84 atm pressure, and their melting points, kanthal—1800°K, and nickelin—1600°K, were above the adiabatic flame temperatures of the limit mixtures. The energy input by the fusing wires could not appreciably affect the initial temperature of the entire volume of gas.

The transition from complete combustion to no reaction occurred directly at the upper limit in the present experiments in agreement with Refs. 2, 5, 6, and 7, and in contrast to the extended transition at the lower limit [6, 7, 18].

Discussion of Results

When an inert gas is added to a hydrogen-oxygen mixture, the oxygen concentration at the upper limit is changed, but the adiabatic-limit flame temperature does not seem to be affected appreciably, as shown in Table 4. Helium, neon, and argon, all of which have smaller specific heats than hydrogen, decrease oxygen concentration, and carbon dioxide, with a higher specific heat, increases oxygen concentration at the upper limit, as shown in Figs. 3–5. However, by addition of equal amounts of inert gases, giving nearly the same adiabatic flame temperatures, the oxygen concentration at the limit at pressures below 5 atm is most decreased by that inert gas which has the smallest thermal conductivity—that is, in the order argon, neon, and helium, as shown in Fig. 4. These sometimes mutually contradicting effects are in agreement with the results of Egerton and Powling [5] and could explain why hydrogen-air at atmospheric pressure gives a lower oxygen concentration at the limit than hydrogen-oxygen. The specific heat of nitrogen is a little higher and the thermal conductivity is considerably smaller than that of hydrogen.

Increase of pressure above that of the atmosphere has a similar effect on the oxygen concentration at the upper limit of flammability of all hydrogen-oxygen-inert mixtures. With increasing pressure, the oxygen concentration at the limit passes through a maximum, which is located at different pressures for compositions of different mixtures. This maximum

Table 4. Adiabatic Flame Temperatures^a in Degrees Kelvin for Upper Limit Hydrogen-Oxygen-Inert Gas Mixtures

Inert	Helium					Neon	Argon	Carbon Dioxide	Air
	0	20	40	60	80	20	20	10	
Vol % Inert									
1.94 atm	1200	1170	1150	1130	1110	1170	1130	1190	1160
9.7 atm	1230	1240	1240	1250	1240	1250	1250	1250	1240

^a Calculated (neglecting dissociation) at constant pressures using gas properties from Ref. 19 with an initial temperature 298°K. A change of 0.1 vol % oxygen corresponds approximately to a change of 20°K.

has earlier been observed by Berl and Werner [7] for hydrogen-air and can possibly be interpreted by the results of Dorr and Schreiner [2] for hydrogen-oxygen mixtures.

The existence of a maximum of the oxygen concentration at the limit could possibly be due to the increasing effect of three-body reactions at elevated pressures. The influence of this change in reaction kinetics upon the equation of conservation of species and energy will be discussed in a subsequent paper.

I wish to thank Professor L. Minnhagen for support and excellent working conditions. I am also indebted to University Engineer H. Lundberg for assistance with the design of the experimental apparatus. It is gratefully acknowledged that this investigation has been supported by the Royal Swedish Navy.

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PAPER II

A LINE HEAT SOURCE AND DETECTOR FOR TRANSIENT
CONVECTION AND IGNITION EXPERIMENTS.

G.S. HOLMSTEDT

DEPARTMENT OF PHYSICS, LUND
INSTITUTE OF TECHNOLOGY, LUND 7,
SWEDEN

Abstract

A line heat source has been developed which is capable of heating fine wires (0.02 - 0.20 mm in diam) to temperatures of up to 2000 K within a millisecond and of keeping the temperature within fixed limits for periods of seconds.

After an initial pulse produced by a thyristor-triggered capacitor discharge, the electrical resistance of the wire is kept constant by means of an electronic feedback system which can handle large variations in cooling-rate and responds within a few milliseconds to changes in the environment of the wire.

Introduction

The steady state natural convection wake from a heated horizontal line source has been studied analytically and experimentally over a long period. A fine wire heated by a constant current or from a constant voltage supply usually serves as the line source [1]. For transient convection experiments in the 1 - 100 msec range [2], the electric current available from such a line source is too limited and, owing to the thermal inertia of the wire, the final temperature is not reached rapidly enough.

The source of ignition most frequently used in flammability tests is an electric spark [3]. The primary advantage of using sparks is that the energy released when discharging a capacitor of known capacitance charged to a known voltage is constant. On the other hand, it is difficult to control many of the factors associated with spark ignition [4]. In order to examine experimentally the growth of an ignition kernel into a combustion wave [5], a line heat source has been developed.

Previous authors have described the static and dynamic properties of some line heat sources. Commercially available CTA (Constant-Temperature Anemometer) bridges and the power supply by Ouwens and Koken [6] are too limited in current to allow the heating of a wire to 2000 K in a millisecond, while the optical feedback system of Preston and Ashman [7] is not easily adaptable to high pressure experiments.

The present heated line source and detector has been found to give a reproducible temperature-time curve in the 1 - 1000 msec range and to respond within a few milliseconds to changes in the environment of the wire. The effect of large variations in cooling rate on the temperature-time performance can be predicted and minor changes in day-to-day atmospheric pressure can be neglected. Proper choice of the length and diameter of the wire keeps the temperature variations within fixed limits.

These properties, which make possible the calibration of the line source in one atmosphere and its use in another, make the line source very suitable for transient

convection and ignition experiments.

Principles of operation

The line source consists of a wire with a temperature-dependent resistance, stretched between two elastic supports. The wire is heated by the current from an electric circuit as shown in Fig 1. During the first millisecond, the wire is heated to its operating temperature T_f by a very reproducible thyristor-triggered capacitor discharge [8]. One millisecond later, the relay 2 connects a power supply which keeps the resistance of the wire at a value corresponding to the operating temperature. The time delay of relay 2 (a Hg-wetted 5 amp reed relay) of about 2 msec and the adjustment of the variable time delay (0.5 - 1.5 msec) determine the time between the initial pulse and the connection of the power supply. This time (about 1 msec) is reproducible to within 10 μ sec using relay 1 (a dual reed relay) as synchronizer.

Apparatus

The power supply in Fig 1 was designed using the feedback system of Ouwens and Koken [6]. For a large gain in the differential amplifier, the supply has a characteristic curve

$$V = V_{\text{ref}} + R_i \cdot I,$$

where $V_{\text{ref}} = V_2 \cdot R_2 / (R_1 + R_2)$ and

$$R_i = R_m \cdot R_1 \cdot R_2 / ((R_1 + R_2) \cdot R_3)$$

By choosing resistors R_1 , R_2 , R_3 and R_m (also used as a current monitor) with a small temperature dependence, a linear characteristic with a standard deviation of 3mA and 3m V was achieved for values of R_i in the range 0-4 ohms and of I from 0-4 amps. The setting of the supply determines the resistance and thereby the temperature of the wire. The influence of temperature-dependent resistors outside the wire was reduced by using thick conductors and supports. The wires were spot-welded and bonded with silver-filled epoxy to the supports. Wires of tungsten were used since these are available with an accuracy of 0.5 o/o in their diameters and have well-known thermal, resistive and light-emissive properties.

Results and discussion

The short initial pulse from the triggered capacitor produces a flat temperature distribution (T_f) along the wire. Owing to cooling at the supports, this distribution changes in time. Since the resistance of the wire is kept at a constant value by the power supply, the temperature at the centre of the wire T will increase to a steady state value T_s . This change from T_f to T_s affects the performance of the line source and has been studied both experimentally and theoretically.

The steady state and the transient temperature of the wire have been studied experimentally for different cooling rates. The temperatures were determined with 0.5 - 1 o/o accuracy, T_f by measuring the wire resistance, T_s and the steady state distribution by using a Cambridge disappearing-filament pyrometer ($\lambda = 0.65 \mu$, $\epsilon_\lambda = 0.45$, window transmisson 0.92). The transient temperature at the centre of the wire, T , was measured using a photomultiplier which had been calibrated in steady state conditions using

the pyrometer. The line source was adjusted as follows: The power supply was connected and by adjusting the potentiometers R_2 and R_3 the characteristic curve which heated the wire to a resistance corresponding to a flat temperature distribution of T_f in the steady state could be selected. The voltage V_3 across the triggered capacitor was then adjusted to heat the wire to T_f . This matching between the triggered capacitor and the power supply could be made accurately by observing the voltage across the wire with a CRO as shown in fig 2. Fig 3 shows the reproducibility of the line source and Fig 4 and in Table 1 the measured performances at different temperatures are compared with theoretical predictions.

The temperature distribution has been determined analytically by solving the time-dependent heat equation when a current $I = V_{ref}/(R - R_i)$ is generated by the power supply, R being the momentary resistance of the wire. A flat temperature distribution was chosen as the initial condition and the boundary supports were assumed to be at a constant temperature of 300 K. Steady state values were used for

the heat lost by convection [9], an approximation valid for gases after a few milliseconds. These calculations which take the temperature dependence of the thermal and resistive properties of tungsten (shown in Fig 4 and Table 1) into account, are in agreement with the experimental results. Calculations for tungsten wires (0.1 mm in diam) with different cooling rates are shown in Fig 5. The temperature difference $T_s - T_f$ depends on the variation of the temperature distribution along the wire with cooling rate and on the power supply. The power supply becomes unstable when the transconductance I/V_{ref} becomes too large. Increasing the length of the wire does not affect the time-scale of the temperature redistribution shown in Table 1, but reduces the temperature differences $T - T_f$ and $T_s - T_f$ by approximately the same factor if the transconductance of the power supply is kept constant. Any mismatch between the triggered capacitor and the power supply is very quickly corrected for by the power supply, as shown in Table 1. The response to sudden changes in cooling rate is shown in Fig. 6. The change in cooling rate is caused by the onset of a chemical reaction. In this way, the line source can be used as a

detector . An estimate of the time constant from CTA theory [10] gives a value of 3 msec with the apparatus used.

Acknowledgments

I wish to thank Professor L. Minnhagen for the provision of laboratory facilities and C. Palm for assistance with the experiment.

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Tables

Table I: calculated and experimental temperatures $T - T_f$ at the centre of a tungsten wire 36 mm in length and 0.1 mm in diameter as a function of the time after discharge. The characteristic of the power supply was unchanged. $T_f = 1300$ K.

Cooling rate atm H_2	$(T - T_f)K$						$(T_s - T_f)$
	0 msec	10 msec	20 msec	30 msec	40 msec	100 msec	K
0.1 cal.	0	25	45	58	68	105	135
0.1 exp.	0	20	43	60	72	111	141
1 cal	0	31	48	59	67	95	112
1 exp	0	30	50	64	78	100	115
1 cal	92	38	48	59	67	95	112
1 exp	56	33	50	63	80	101	116
10 cal	0	35	49	58	65	85	90
100 cal	0	35	47	54	59	69	71

Figure captions

Fig 1. The heated line source. V_1 V_2 and V_3 - voltages from stabilized dc supplies. D - differential amplifier.

Fig 2. The matching between the triggered capacitor and the power supply for different capacitor voltages V_3 . Blurred trace (negative pulse) - T measured by a photomultiplier. Distinct trace - wire voltage.

Fig 3. Two exposures of the line source taken at an interval of 1 hour with an unchanged setting. Lower trace (negative pulse) - T. Upper trace - wire voltage.

Fig 4. Calculated (-) and measured (0.1 atm, x 1 atm) temperature distributions along a tungsten wire 36 mm in length and 0.1 mm in diameter.

Fig 5. Steady state current - voltage - temperature curves for a tungsten wire 36 mm in length and 0.1 atm in diam. Straight solid lines (-) T_s . Lines through the origin and the points marked x(.....) - T_f .

Fig 6. Ignition of a $H_2 - O_2$ mixture by a hot wire.

Upper trace (negative pulse) - T.

Lower trace-wire voltage.

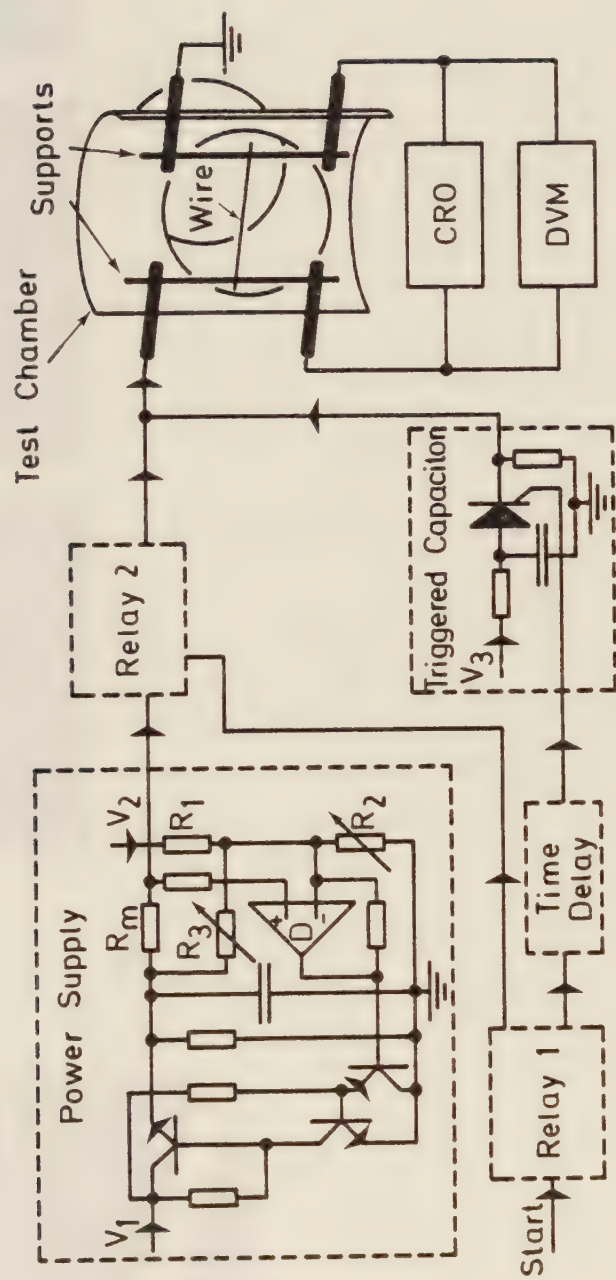


Fig 1

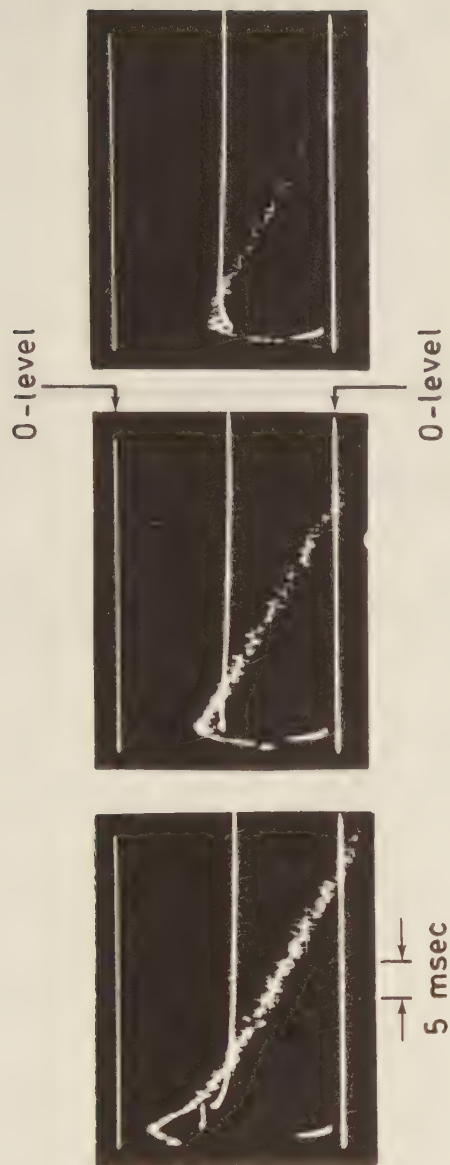


Fig 2

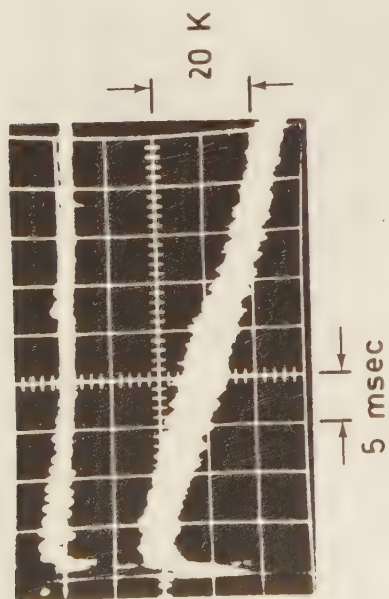


Fig 3

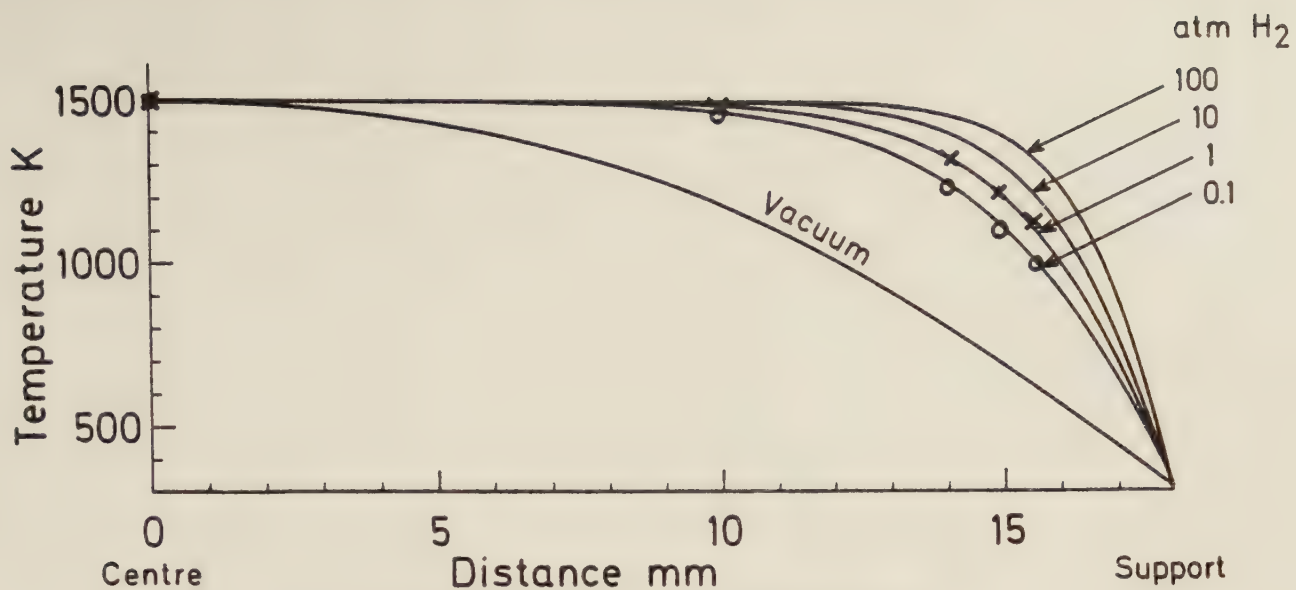


Fig 4

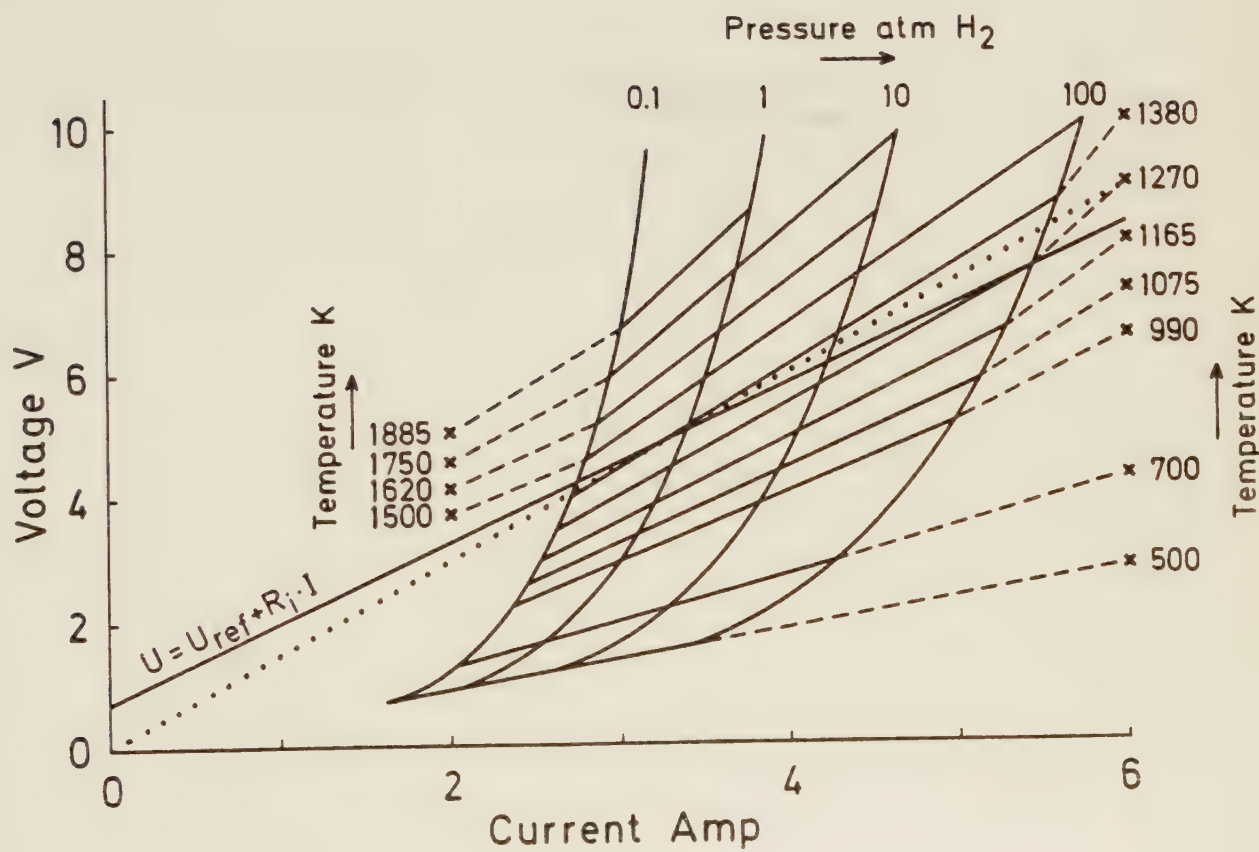


Fig 5

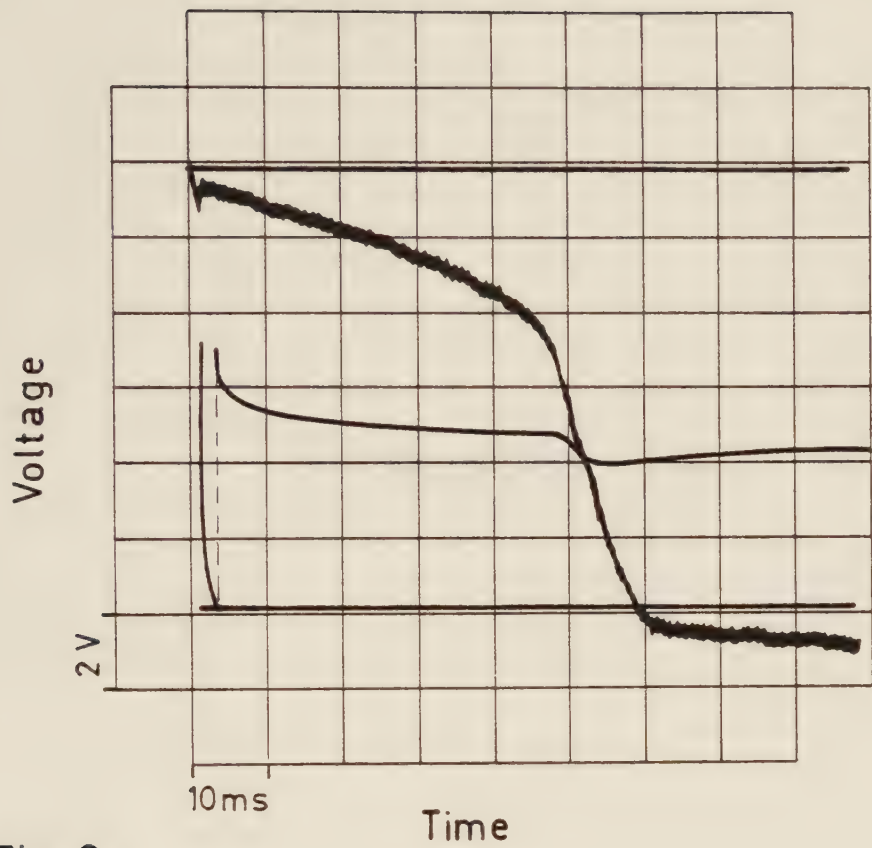


Fig 6

PAPER III

EXPERIMENTAL INVESTIGATION OF THE ONSET OF FREE
CONVECTION NEAR A SUDDENLY HEATED HORIZONTAL WIRE.

G.S. HOLMSTEDT

DEPARTMENT OF PHYSICS, LUND

INSTITUTE OF TECHNOLOGY, LUND 7

SWEDEN

Abstract

The temperature field in a fluid surrounding a horizontal wire which is suddenly heated has been studied with a Mach-Zehnder interferometer. It has been found experimentally that the outer isotherms start to move upwards immediately with a constant acceleration for about 100 msec. After 300-400 msec, the velocity becomes constant. A simple model predicts the acceleration as a function of the energy dissipation in the wire and the transport properties in the fluid. Experimental data with different gases, pressures and wire diameters are in substantial agreement with the theoretical predictions.

Nomenclature

A	acceleration
c_p	specific heat at constant pressure
d	diameter of the wire
g	acceleration due to gravity
k	thermal conductivity
L	length of the wire
n	distance ratio parameter
Nu	Nusselt number
q	heating rate per meter
r	distance from the wire to an isotherm
Ra	Rayleigh number
t	time
T	temperature K
x	vertical displacement
σ	Nusselt number ratio parameter
ρ	fluid density
λ	wavelength of laser light

Subscripts

∞	without influence of the wire
air	air property
f	film value derived at $(T_w - T_\infty)/2$
h	horizontal
H_2	hydrogen property
m	mean value inside an isotherm
N_2	nitrogen property
v	vertical
w	wire property

1 Introduction

The onset of convection near a suddenly heated surface is of importance in many contexts including the formation of thermals and plumes [1,2,3,4] , the use of hot wire anemometers [5] and the ignition of gas mixtures [6] .

Most theoretical studies of this subject start from the Boussinesq approximation. The solution for small times and large Grashof numbers using the boundary-layer approximation is given by Elliot [7] for an axisymmetric body whose temperature is suddenly increased. Gupta and Pop [8] have considered the influence of curvature effects during the initial period. These solutions, however, only correspond to physical phenomena for sufficiently large Grashof numbers. For lower values of Grashof numbers, there have been very few theoretical studies. A model which explains the generation of thermals has been proposed by Howard [9]. He postulates that when the Rayleigh number, Ra , based on the instantaneous thickness of the conduction layer, reaches a critical value, the layer will break up and form a thermal. The critical value of Rayleigh number $Ra = 1100$ is taken from

the solution of the classical Benard problem (an infinite fluid layer above a horizontal surface). This model has been used by West and Lawson [10] to explain their experimental results on the onset of natural convection. They found a delay time between the application of power to the line source and the beginning of observable convection. In Refs [2,11,12,13], some aspects on the formation, growth and motion of laminar thermals are described.

There appears not to have been any detailed work on the onset of natural convection exploring the interval between the start of heating and the plume stage with a constant velocity.

The present study investigates the onset of natural convection above a line source in gases with source temperatures of up to 1400 K, a region of considerable interest in ignition studies [14] which is far away from the range of validity of the Boussinesq approximation [15].

2 Injection methods

Different methods have been used to inject density gradients into a fluid. In experiments with steady state natural convection [13,16,17,18], the fluid is usually brought into contact with a hot solid surface. In experiments with transient convection in liquids, electro-chemical [3,19] and single-electrode conductivity [1,2] sources have also been used to determine accurate values of injection rates and reduce the influence of the thermal inertia of the source. In gases where these methods are not applicable, electrical heating of wires is most commonly used [11,12,13].

Theoretical models usually assume a source with a constant heating rate or with a step change in the temperature of its surface. A constant heating rate is easily achieved by passing a constant current through a wire having a resistance with a small temperature-dependence. Such sources, however, have long time-constants. Solving the equation for the conservation of energy in a long wire assuming a constant value of $Nu_f \times k_f$ as an approximation, shows

that a fraction $e^{- (Nu_f \times k_f / (Q_w \times c_{p_w} \times d^2) \times t)}$

of the heating rate remains in the wire.

Thus, the time scale for the experiment must be

several times $t = 3Q_w \times c_{p_w} \times d^2 / (Nu_f \times k_f)$ which is

the time when 5 o/o of the heating rate remains

(and 32 o/o of the heat input is stored) in the wire.

This condition restricts the use of simple,

constant power sources in gases to experimental

times above 1 sec unless wires with diameters

of a few microns are used.

On the other hand sources producing step changes' in the temperature are difficult to make.

Commercially available constant-temperature anemometers, using thin wires with a temperature-dependent resistance, having a very small time constant [20], are too limited in current for

wires 20-200 μm in diam. A constant temperature source has been developed for these experiments [14].

This source heats a wire, 20-200 μm in diam, to a temperature of up to 2000 K within a milli-second and keeps the temperature within fixed limits for periods of seconds.

3 Apparatus

The apparatus used is shown in Fig 1. The experiments were conducted in a brass vessel 10 cm in diam and 13 cm in length in H_2 and N_2 atmospheres and in a rectangular box 20 x 30 x 25 cm in air. Heat was injected into the gas through hot tungsten wires, 50, 100, and 200 μm in diam having $L/d = 700 - 1800$. The wires were stretched between two elastic copper supports. By rotating the test chamber with two adjustment screws, the wire could be aligned horizontally in the collimated light beam after the vessel was sealed.

The resistance and hence the temperature of the wire were kept constant by means of an electronic feedback system consisting of a combination of a thyristor-triggered capacitor discharge and a power supply [14]. In this way, temperature fluctuations during an experiment could be kept to within 2 o/o of the operation temperature T K. 1.5 o/o of the fluctuations were due to a slow reproducible increase in the temperature due to the redistribution of heat along the finite wire and 0.5 o/o was due to the calibration procedure.

The latter was performed in steady state conditions using a Cambridge disappearing-filament pyrometer ($\lambda = 0.65 \mu\text{m}$, $\xi_\lambda = 0.45$, window transmission 0.92) and the known temperature-dependence of the resistance.

Transient temperature distributions in the fluids were obtained using a Mach-Zehnder interferometer 10 cm in diam and high-speed photographic equipment. The specially-designed interferometer was built on a 20 mm aluminium plate resting on foam rubber on a vibration-proof table. The optics of the entire interferometer were flat to within $1/10 \lambda$ and the test and compensating windows to within $1/20 \lambda$. The interferometric translation and rotation stages allowed adjustment in such a way that only one colour appeared in the test field, the adjustment being stable over a period of one day.

Two different light sources were used, a tungsten lamp for the adjustment of infinite fringe width and a pulsed, stacked-diode laser

(RCA SG 3001) for high-speed photographic recording. The infrared diode laser ($\lambda = 0.904 \text{ } \mu\text{m}$) is well suited for high-speed recordings with interferometers and schlieren equipments since it has a small light-emitting area ($0.2 \times 0.2 \text{ mm}$) and a high peak radiant flux (40 W). A pulse duration of $0.2 \text{ } \mu\text{sec}$ giving an energy in the pulse large enough for photographic registration, permits the laser to be used at a repetition rate of 2 kHz at room temperature and of 80 kHz with the laser cooled to cryogenic temperatures. The spectral width of the diode laser is small enough to give more than 200 fringes but large enough to avoid unwanted diffraction and interference patterns from objects out of focus. The infrared light can easily be made visible with a Kodak JR phosphor.

Photos were taken on $24 \times 36 \text{ mm}$ Kodak high speed infrared film with a Nikon Nikomat camera. A $300 \text{ mm}(f 1: 4.5)$ objective mounted on bellows reduced the picture format by a factor 2-3. The rear part of the camera was replaced by an electromechanical device feeding the film with velocities up to 20 m/s . The edges of the

film were exposed with two infrared emitting diodes, one synchronized with a 1kHz standard making 1 msec ticks, the other indicating the start of the measurement.

The interferograms were analysed with a Zeiss Abbe comparator equipped with a photoelectric setting device.

4 Results and discussion

4.1 Evaluation of the interferograms.

The temperature field in the fluid was determined as a function of time for several sets of working conditions: wire diams between 50 - 200 μm , temperatures $T_w - T_\infty$ between 50-1100 K, pressures between 0.5 and 3 atm and the gases H_2 , N_2 and air.

In Fig 2 some interferograms, usually taken every two msec, are shown. Using the comparator with the photoelectric setting device, the horizontal and vertical distances r from the wire to the points corresponding to the maxima and minima in the optical density (differing by $\lambda/2$ in optical path length) were determined.

In Fig 3 (which corresponds to Fig 2 a), the different stages in the displacement of a typical isotherm are shown: a conduction and acceleration stage (0 - 300 msec) and a constant velocity (plume) stage. These investigations consider that part of the conduction and acceleration stages (≤ 60 msec) where the displacements x are much smaller than the distance r of the outer isotherms.

4.2 Thermal diffusion.

Measurements of the distance from the wire to the horizontal isotherm r_h can be related to pure thermal conduction since the vertical displacement x of the temperature field during the initial period is small. A plot of r_h^2 against t gives a straight line after a few milliseconds as shown in Fig 4, the value of r_h being reproducible to within a distance corresponding to an optical path length difference of $\lambda/10$. The correction due to the finite wire length is below $\lambda/30$.

The r^2 -- t dependence observed is also predicted by the theory of conduction in solids using a constant line source [21]. To compare the experimental findings for compressible fluids having temperature-dependant physical properties with theory, a numerical method has been used.

The coupled continuity, momentum and energy equations for an axisymmetric body, as given by Pai [22], were solved using an explicit finite difference method, but neglecting the force of gravity. Starting from the initial conditions at time $t=0$, the velocity, temperature and density fields at time $t + \Delta t$ were obtained for the given boundary conditions. In order to check the convergence of the finite difference solutions, the spatial mesh sizes were doubled - this was accompanied by an appropriate change in the value of Δt - and results for the two solutions compared. The maximum difference observed was found to be 5 o/o in dependent variables and this was considered to be an acceptable degree of convergence. The convergence requirement made it necessary to use very small spatial mesh sizes near the wire. Thus, in order to keep the computing times within reasonable limits, two spatial mesh sizes were used. It was found that when the initial pressure wave, produced by the step change in the temperature of the wire, had left the temperature field, the solution was controlled by conduction, viscous effects being negligible. Hence, the main part of the calculations was performed using only the continuity and energy equations. From the solutions

for different gases (N_2 , H_2), pressures (1-5atms) and temperatures $T_w - T_\infty$ (200-1100K) the following conclusions can be reached:

a) After an initial rapid decrease, the density gradient and hence the heating rate decrease slowly with time (by less than 20 o/o between 5 and 40 msec after the application of heat to the wire).

b) The average density inside an isotherm ρ_m is constant in time (the variations being less than 2 o/o between $t=5$ and $t=40$ msec) and, for a given value of $T_w - T_\infty$ and d , ρ_m/ρ_∞ is identical for different gases (the variations are less than 2 o/o for H_2 and N_2).

c) The correlation between $(T - T_\infty)/q$ and $r^2 \cdot \rho_\infty \cdot c_p / k_f \cdot t$ (shown in Fig 5) indicates that a $r^2 - t$ dependence also holds for compressible fluids, in agreement with the experimental findings.

The $(T - T_\infty)/q$ values for the hydrogen in Fig 5 have been multiplied by a factor 4.5 to facilitate a comparison between the isotherms of H_2 and N_2 . For given values of $T_w - T_\infty$, d and pressure, $q_{H_2}/q_{N_2} \approx Nu_{H_2} \cdot k_{f,H_2} / Nu_{N_2} \cdot k_{f,N_2} = 4.5$ to within 5 o/o for the ranges of $T_w - T_\infty$, d and pressure examined.

4.3 Acceleration stage.

The vertical displacement of the temperature field was determined by accurate measurements of the distances r from the wire to isotherms as a function of time. In Fig 6, r is shown as a function of time for a typical outer isotherm. It is found that the vertical shift x of isotherms above and below the wire are the same and are proportional to t^2 . This indicates a constant acceleration A as long as x is small compared to r . The variation in the measured acceleration between outer isotherms was below 10 o/o. In Fig 7, the acceleration (usually the mean value of the $\lambda/2$, λ and $3\lambda/2$ isotherms) is shown to be a linear function of $T_w - T_\infty$ for different gases at atmospheric pressure with wires 100 μm in diam. Disconnecting the triggered capacitor, which results in a line source with a much longer time-constant, gives the same results as the introduction of a delay-time as shown in Fig 7. This is probably the reason for the discrepancy between the findings in the present work and those by Ref. 10 performed in identical conditions in air.

In solving the equations for an instantaneous point source of heat, using spherical polar coordinates and the Boussinesq approximation,

Morton [23] has shown r^2-t to be a necessary condition for similarity. Since it can be shown that a corresponding solution cannot be found for a continuous line source and since in addition the Boussinesq approximation is not valid for the temperature range used in these experiments, a simple model to explain the principal features of the experimental findings has been proposed. The model, already used to some extent in Ref. , considers the mass per unit length contained inside an outer isotherm, having $r=n \cdot \sqrt{k_f \cdot t / \rho_\infty \cdot c_p}$. Here, n is the only parameter, since as shown above ρ_m (and the corresponding T_m) and q have constant values and ρ_m / ρ_∞ is independent of the gas used. Neglecting the viscous resistance in the momentum equation leads to:

$$\frac{\delta}{\delta t} [\pi \cdot r^2 \cdot \rho_m \frac{\delta x}{\delta t}] = g \cdot \pi \cdot r^2 \cdot [\rho_\infty - \rho_m]$$

and, after integration,

$$x = (g \cdot [\rho_\infty - \rho_m] / 4 \cdot \rho_m) \cdot t^2$$

By introducing the added heat $q \cdot t = \pi r^2 \rho_m c_p [T_m - T_\infty]$ the acceleration can be expressed as a function of the heating rate:

$$\text{Eqv.1} \quad A = g \cdot q \cdot \rho_\infty / [\rho_m \pi^2 \cdot 2 \cdot \pi \cdot k_f \cdot T_\infty]$$

or, as a function of $T_w - T_\infty$, by approximating the heating rate with the steady state value

$$q = \pi \cdot k_f \cdot \text{Nu} \cdot (T_w - T_\infty) :$$

$$\text{Eqv.2} \quad A = g \cdot \text{Nu} \cdot \rho_\infty \cdot [T_w - T_\infty] / [2 \cdot \pi^2 \cdot T_\infty \cdot \rho_m]$$

The parameter n , often used as a measure of the width of the conduction layer [1,3,10,11], depends on the properties of the gas. Comparing Eqv.2 with the results shown in Fig 7. gives a constant value of n , since Nu has a slow decrease and ρ_∞/ρ_m a slow increase with $T_w - T_\infty$. The n values for the wire, 100 μm in diam, are $n_{H_2} = 3.1$, $n_{air} = n_{N_2} = 3.8$ and $(n_{N_2}/n_{H_2})^2 = 1.5$. The correlation given in Fig 5 shows $n_{N_2}^2$ to be larger than $n_{H_2}^2$ for all isotherms. A comparison between the added heat, using the steady state value for the heating rate and assuming that ρ_m/ρ_∞ is independent of the gas used, gives $(n_{N_2}/n_{H_2})^2 = Nu_{N_2}/Nu_{H_2}$, which is equal to the experimental value 1.5 for a wide range of wire diameters (20-200 μm) and temperatures ($T_w - T_\infty$) (200-1100K). In Fig 8 the measured values of acceleration, using different wire diameters, gases, pressures and heating rates (experimental steady state values) are shown to be in agreement with the predictions of Eqv.1. Thus, the influence of convection currents near the line source during the acceleration stage can be minimized by accelerating the source with an acceleration given by Eqv.1 [14].

Acknowledgments

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Figure captions

Fig 1. Experimental equipment

A - diode-laser, B - tungsten lamp , C - Mach - Zehnder interferometer , D - compensation chamber, E - test chamber , F - wire , G - photomultiplier tube , H - high-speed photographic recording system.

Fig 2. Interferogram.

Fig 3. Distances to an isotherm as a function of time. The isotherm chosen corresponds to the outermost white fringe in Fig 2a.

Fig 4. Horizontal (displacement)² of some isotherms ($\lambda/2$ apart) as a function of time. $T_w - T_\infty = 800$ K, 1 atm pressure. • - expr nr Int 4, — - Int 15, | - Int 16. Solid lines — theoretical predictions.

Fig 5. Theoretical correlation of T/q as a function of $\text{Log}(r_\infty^2 \cdot c_p / (t \cdot k_f))$

Fig 6. Displacement of an outer isotherm as a function of time. $T_w - T_\infty = 1100$, H_2 at 1 atm pressure. Solid line — constant $\cdot t^2$.

Fig 7. Acceleration of the outer isotherm as a function of $T_w - T_\infty$ for a wire 100 μm in diam.

- - H_2 ; ↓ - air; ← - N_2 .
- ⊙ - 1 - triggered capacitor energy 10 o/o too high.
- ⊙ - 4, 3 and 2 accelerations measured after 20, 30 and 45 msec in an experiment without the triggered capacitor.

Fig 8. Acceleration of the outer isotherms as a function of $g \cdot q / (2 \cdot \pi \cdot k_f \cdot T_\infty \cdot \sigma)$ at different pressures and wire diameters ($\sigma_{\text{H}_2} = 1, \sigma_{\text{N}_2} = 1.5$). $T_\infty = 293 \text{ K}$

- - 1 atm, 100 μm ; ← - 0.53, 1, 47, 2, 9 atm, 100 μm ;
- ↓ - 1 atm 50 and 200 μm .

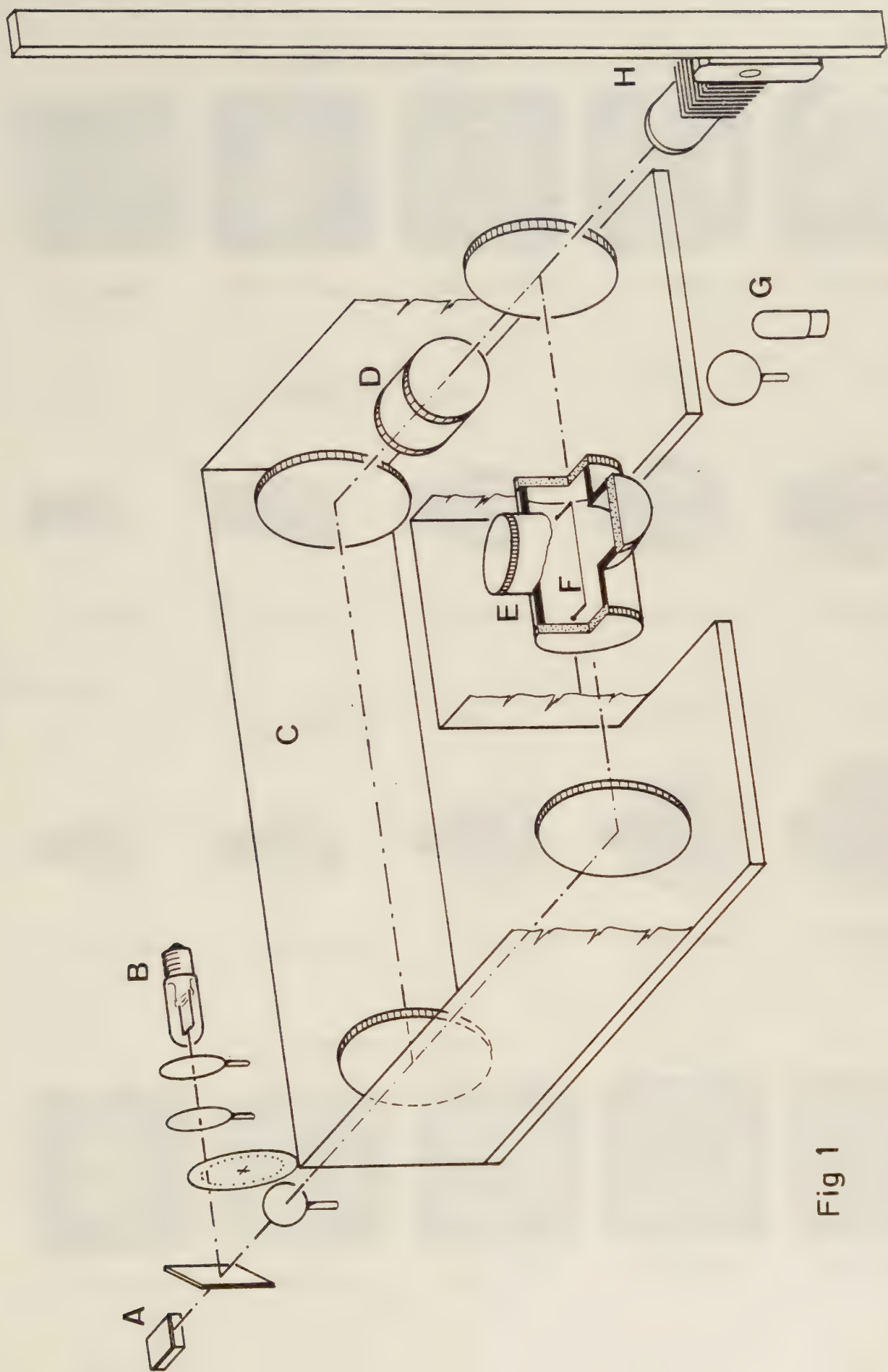


Fig 1

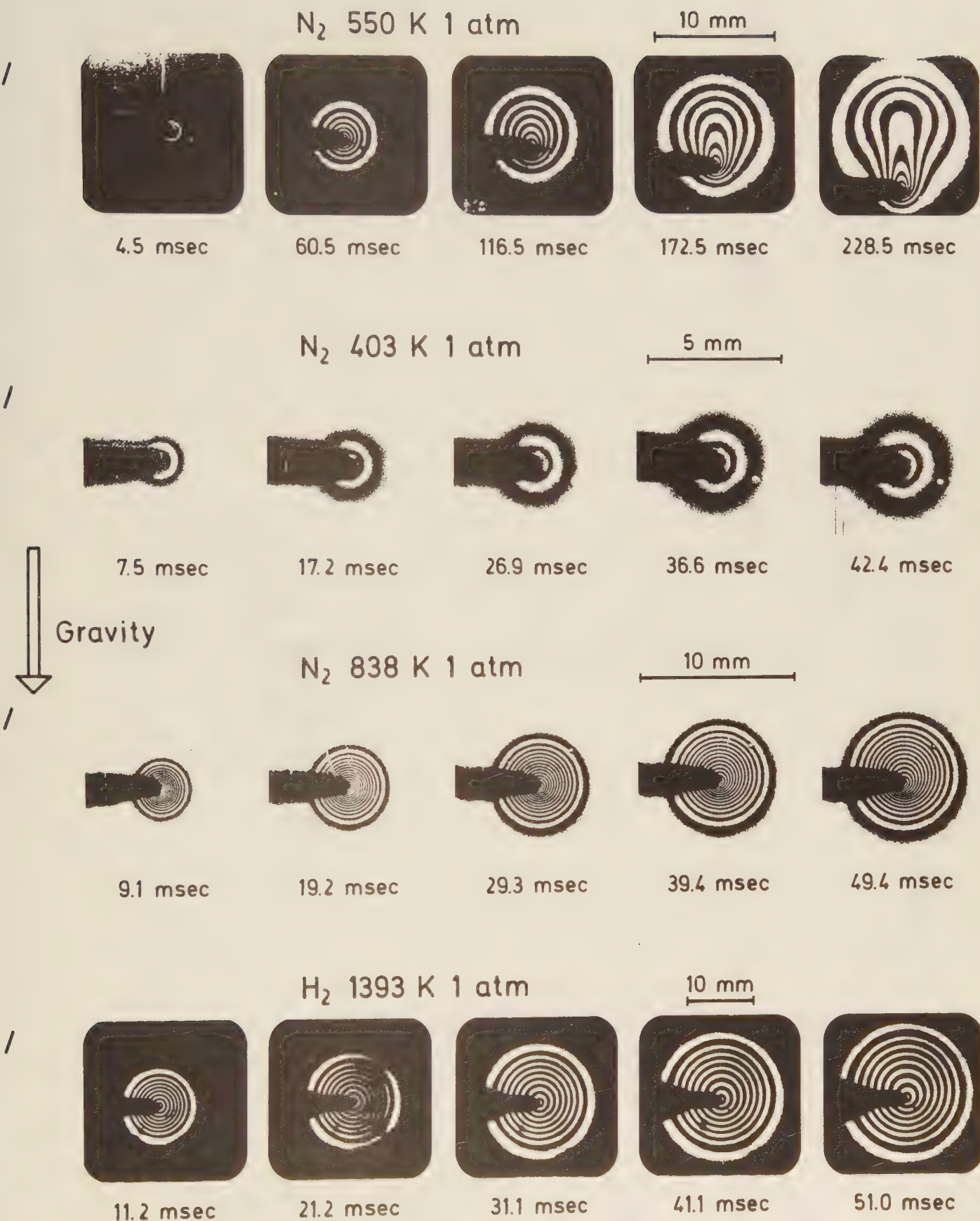


Fig 2

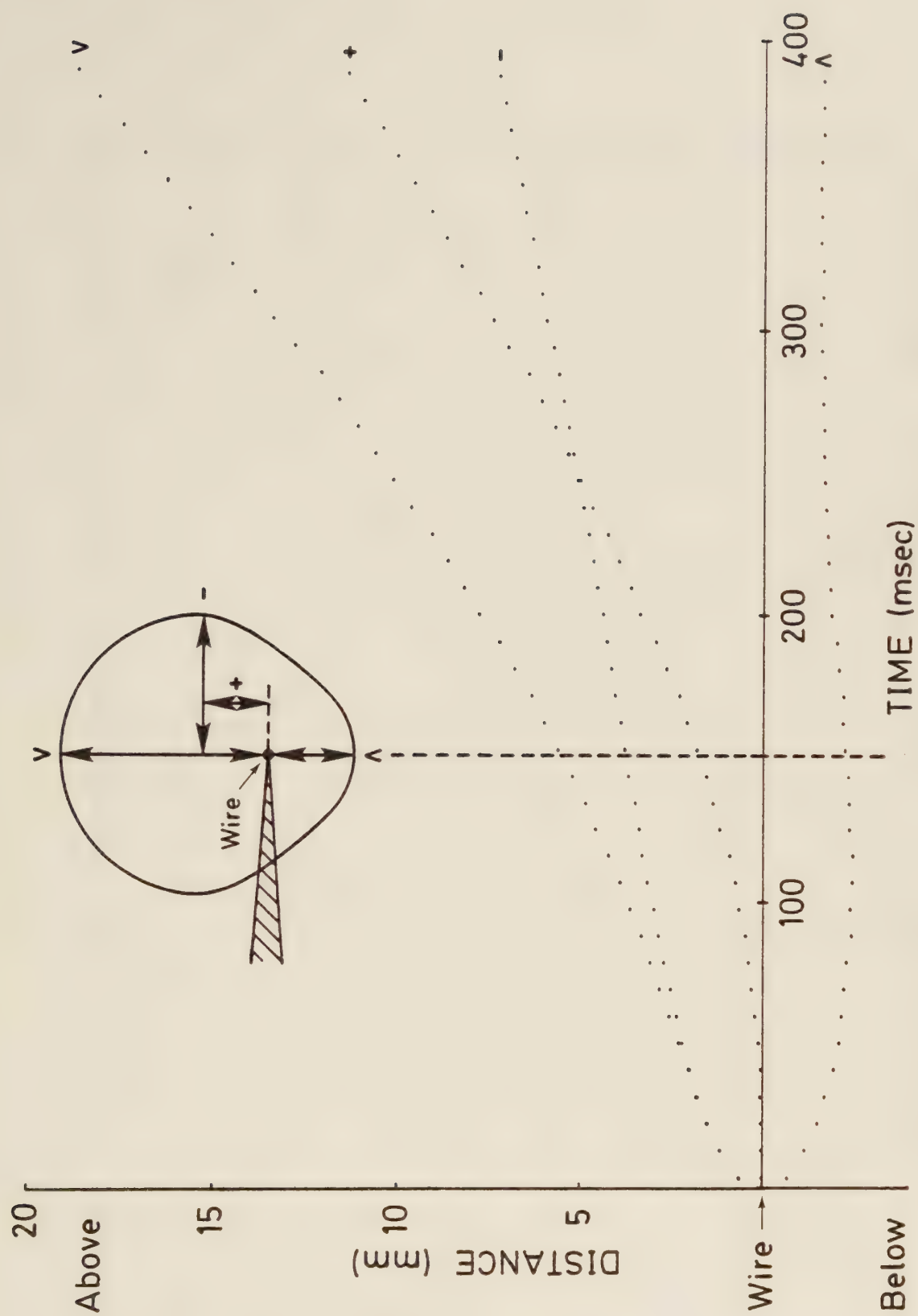


Fig 3

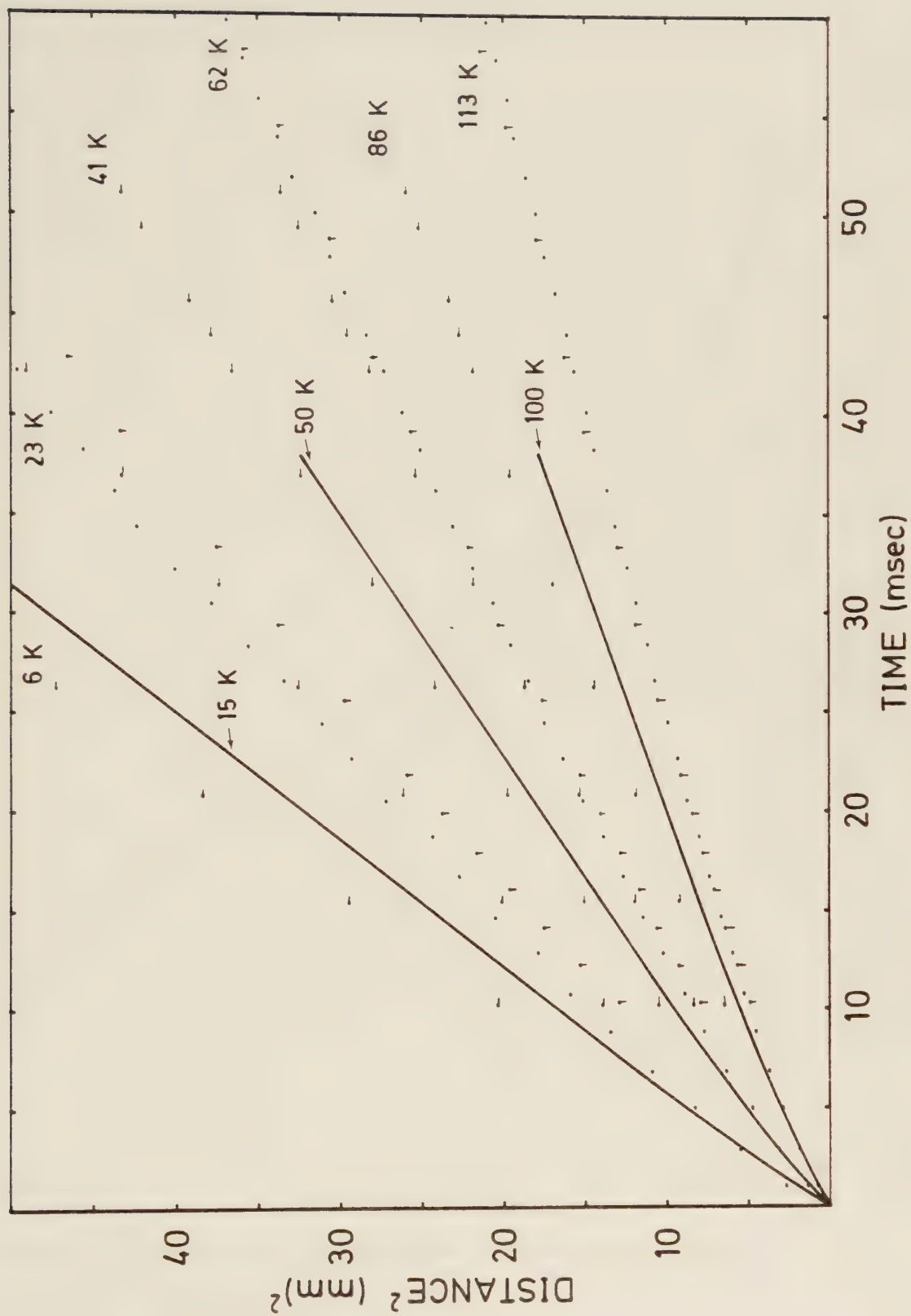


Fig 4

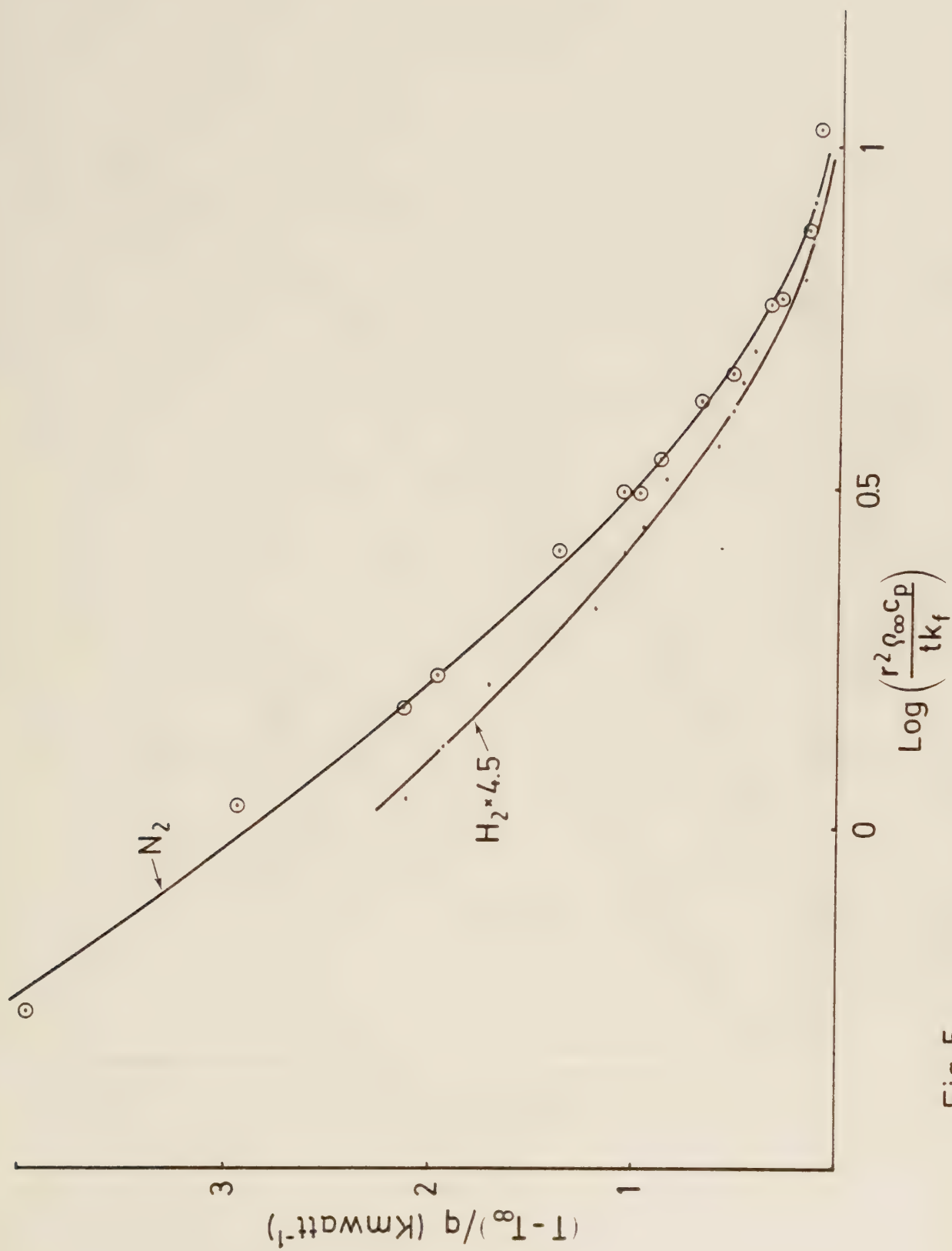


Fig 5

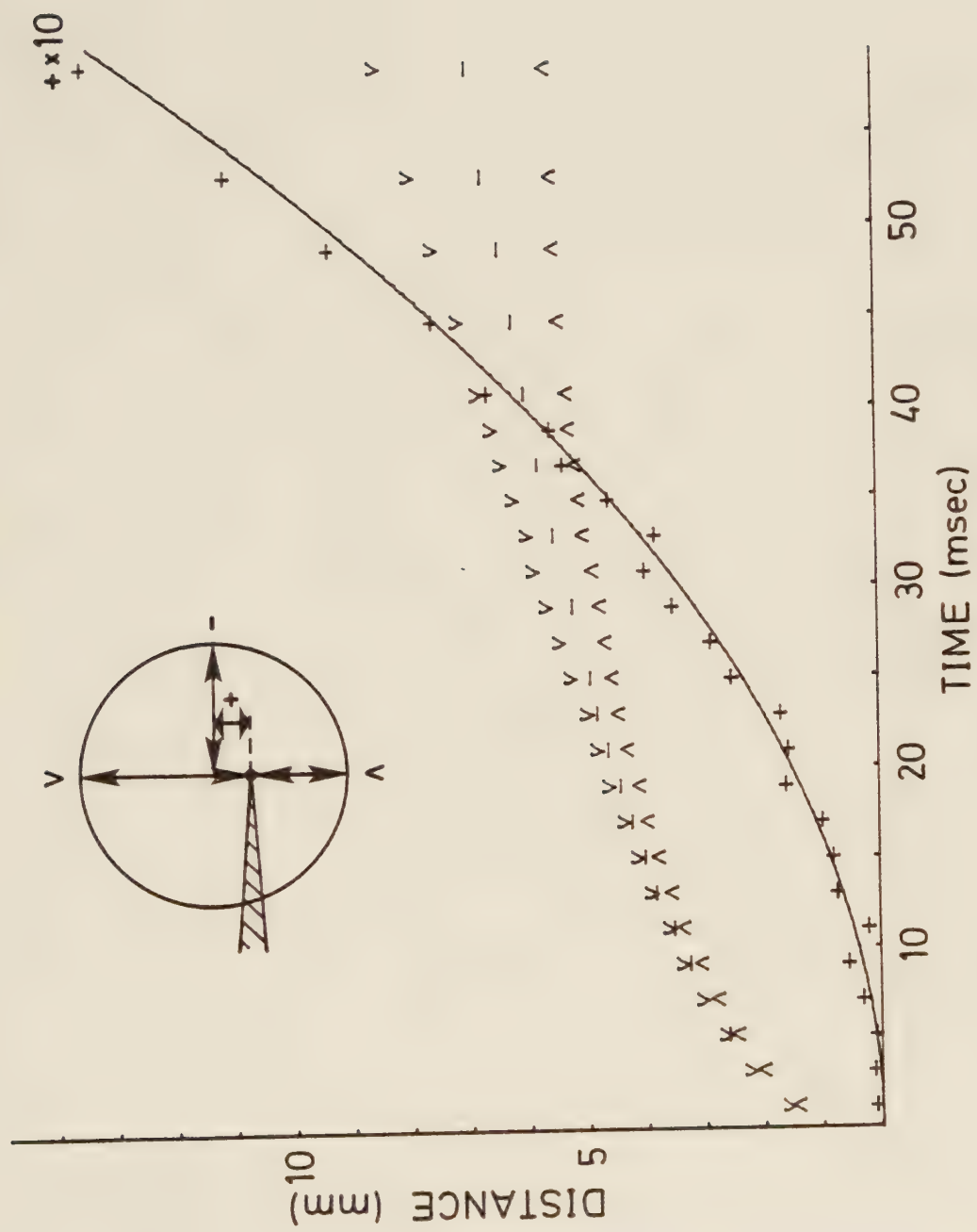


Fig 6

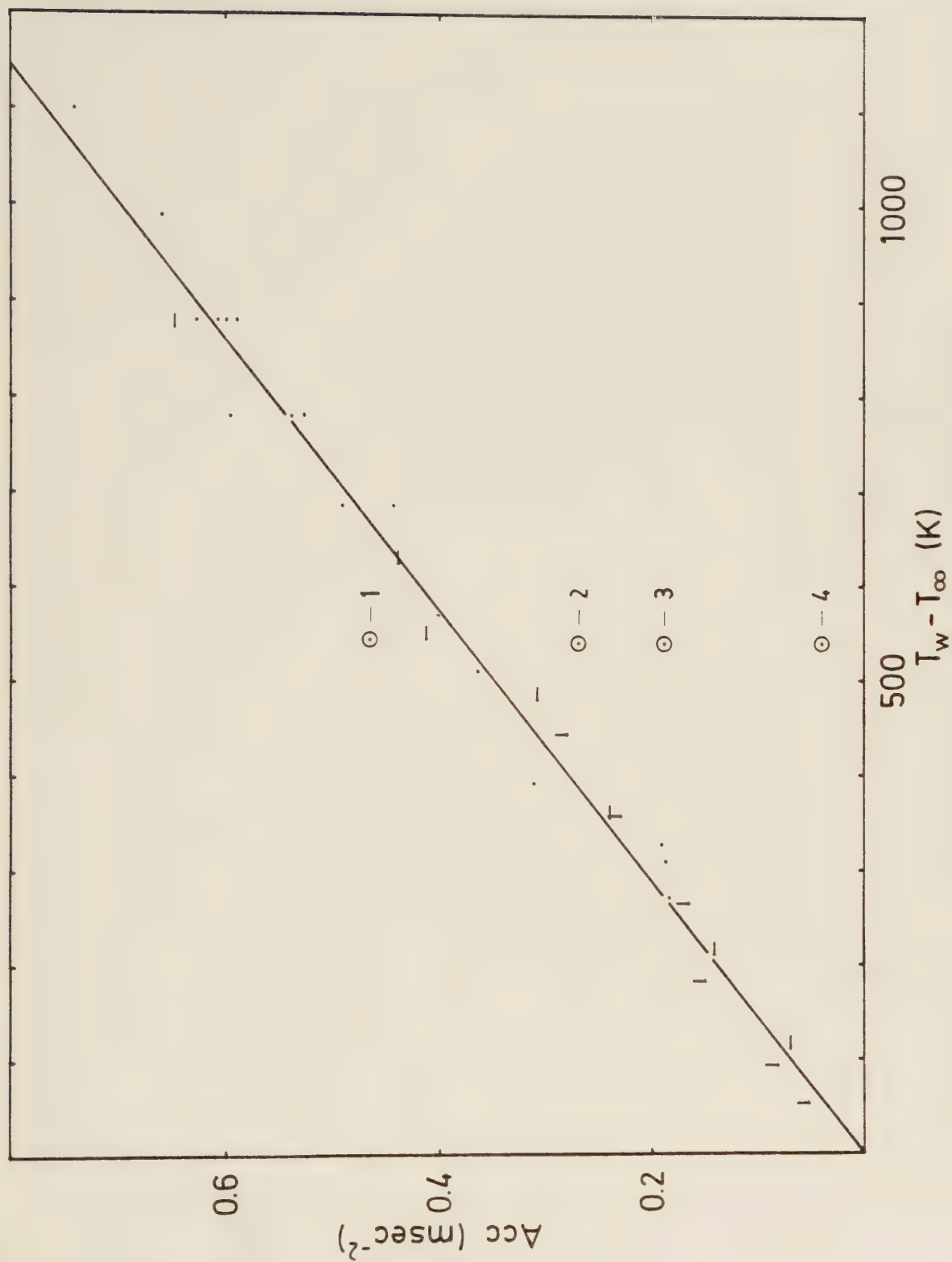


Fig 7

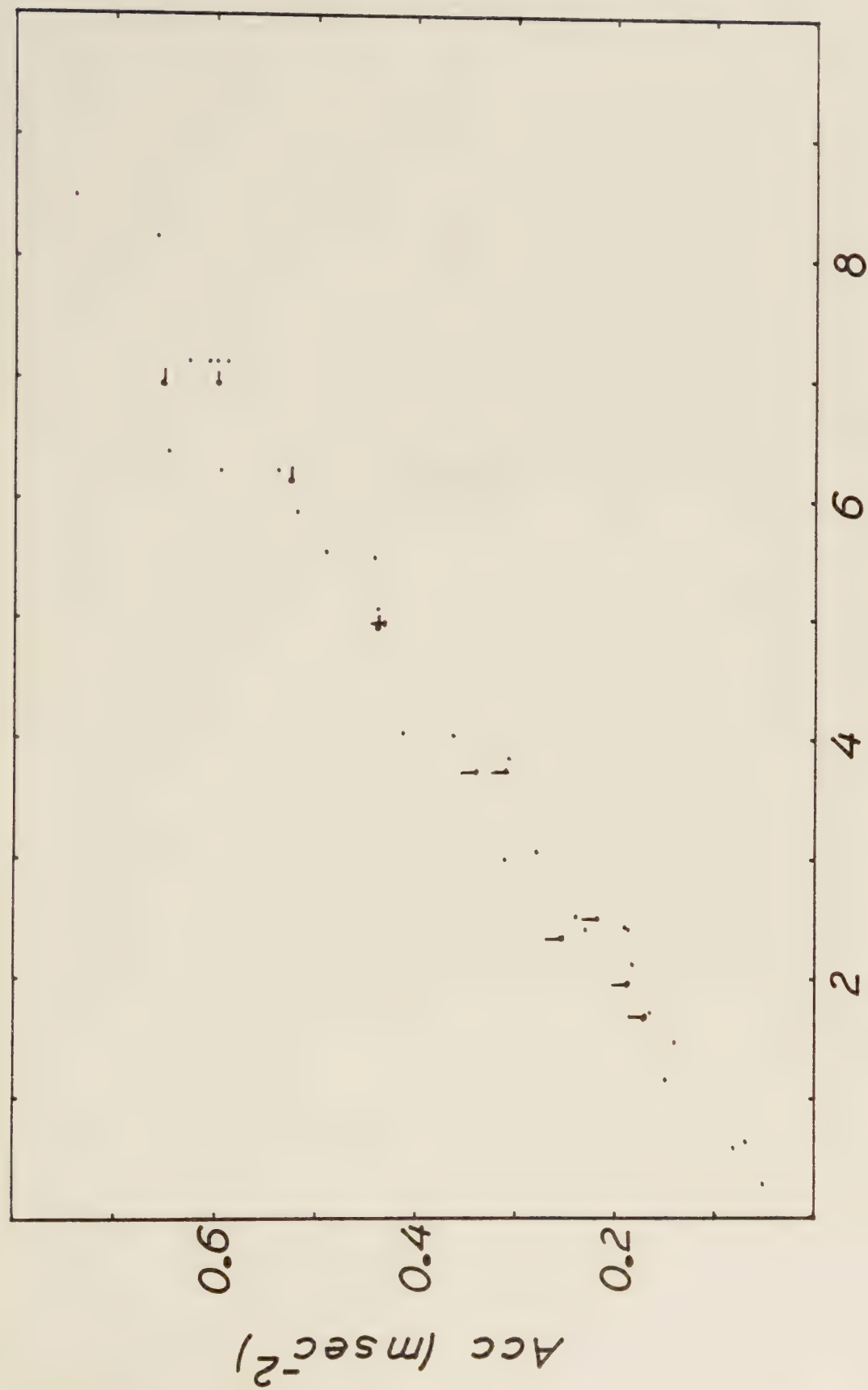


Fig 8 $g \cdot q / (2 \cdot \pi \cdot T_{\infty} \cdot k_f \cdot \sigma) \text{ (msec}^{-2}\text{)}$

PAPER IV

THE IGNITION OF REACTIVE GASES NEAR THE FLAMMABILITY
LIMITS BY HEATED WIRES.

G.S. HOLMSTEDT

DEPARTMENT OF PHYSICS, LUND

INSTITUTE OF TECHNOLOGY, LUND 7.

Abstract

Measurements of burning velocities and of the ignition of hydrogen-oxygen mixtures near the upper limit of flammability in a closed vessel are reported. Close to the limit, ignition is affected by convection currents and is preceded by a reaction zone around the wire, which remains after the limit is passed.

Introduction

Between certain concentration limits, a flame can propagate through a combustible premixed gas at temperatures below the point where spontaneous ignition can occur, if energy is supplied from an external ignition source. Ignition takes place after an "ignition delay". Most frequently electric sparks [1] have been used as a source of ignition and minimum ignition energies and quenching distances to be used in applications or for safety reasons have been determined. Electrodeless sparks produced by a focused laser beam [2] have been used in order to eliminate the quenching influence of the electrodes and hot metal surfaces are commonly used in experiments in which the limits of flammability are to be determined [3,4,5]. Shortly after ignition in large vessels, the flame usually grows spherically [6] if the composition of the mixture is not too close to the limits of flammability. As the limit is approached from the flammable range, both the ignition energy and the quenching distance increase rapidly 1 as does the ignition delay if a hot surface with a constant temperature is used. After ignition, the spherical flame will be deformed 6 when the flame velocity becomes so small that convection currents have time to develop [6]. Thus , special attention has to be paid to the design of an ignition source determining the limits of flammability and the threshold temperatures of hot surfaces. If a source of too low energy is used, the ignition limit might be taken for the limit of flammability, while using one of too high

energy in relation to the test volume can result in partial combustion, although a flame cannot propagate in the gas. The present experiments study the ignition of near limit hydrogen-oxygen gas mixtures with a hot wire in a closed vessel in an attempt to compare different criteria used to define the limits of flammability.

Apparatus

The apparatus used is shown diagrammatically in Fig 1. The experiments were conducted in a cylindrical brass vessel 10 cm in diameter and 13 cm in length, provided with a bursting-diaphragm to protect the two windows of interferometric quality mounted on the end faces. The gas mixtures (gases of 99.99 vol o/o purity) were prepared in the vessel itself by the method of partial pressures using a mercury manometer with an accuracy of 0.2 o/o. The compositions of the mixtures were analyzed with a gas chromatograph (Varian 90 P4) equipped with a thermal detector, an integrator with digital read-out and a high precision sample loop. By switching between the test mixture and a reference mixture (accurate to 0.01 vol o/o) the concentrations of the binary gas mixtures could be determined to 0.05 vol o/o accuracy throughout the oxygen range used (4-7 vol o/o). In all experiments, the measured nitrogen content due to incomplete evacuation before mixing was below 0.05 vol o/o. The gas mixtures were ignited by hot tungsten wires 100 μ m in diameter and 36 and 72 mm in length. A specially-designed line

source has been developed for these experiments [3]. The source, consisting of the combination of a thyristor-triggered capacitor discharge and a power supply, heats the wire to a temperature of up to 2000 K within a millisecond and keeps the temperature fixed to within 2-4 o/o depending on the length of the wire. Of this variation, 0.5 % is due to the calibration procedure and 1.5-3.5 % to temperature redistribution along the finite wire.

The source can also be used to detect the ignition delays since it responds rapidly to changes in the heating rate [3].

The wires were spot-welded and bonded with silver-filled epoxy to the elastic supports. The supports were mounted on a shaft, aligned parallel to the wire, this being attached to the vessel with ball bearings. A constant force could be applied by an electromagnet to the support. Its time constant was less than 0.5 msec and hence the wire could within a millisecond be given a constant vertical acceleration. The acceleration, monitored by light deflection, could be varied by changing the mass of the supports or the voltage to the electromagnet. The development of the ignition process was observed with a Schlieren mirror system perpendicular to the wires and a Mach-Zehnder interferometer parallel to the wires together with a high-speed photographic recording system. The specially designed interferometer and photographic recording system using an exposure time of 0.2 usec and repetition rates of up to 2 K Hz is described in Ref 3 .

Results

The experiments were carried out with hydrogen-oxygen mixtures with compositions near the upper limit of flammability at an initial temperature of $20 \pm 1^\circ\text{C}$ and at atmospheric pressure. The wires, 36 mm in length for Schlieren pictures and 72 mm for interferograms, were heated to temperatures between 1200K (approximately the adiabatic flame temperature at the upper limit of flammability[3]) and 1450K. Preliminary test showed that once a wire had been used several times in successful ignition tests, the temperature distribution along it was no longer smooth due to evaporation of the material. It was impossible to get reproducible results using such wires and hence the wires were changed after every successful ignition.

In Figs 2 and 3 are shown some typical Schlieren pictures and interferograms. Outer fringes in the interferogram at points where reactions do not occur correspond to a certain mean density along the light beam. As shown by the Schlieren pictures, the curvature of the flame in a plane parallel to the wire is not so large and hence the distance to an outer fringe can be taken as a rough measure of the advance of the flame. In Fig 4 the measured distance from the wire (1425K) to the outermost fringe below it is shown as a function of time for mixtures at atmospheric pressure near the upper limit of flammability. Three ranges of composition may be defined on the basis of different flame behaviour: I. For an oxygen content greater than

about 5.5 vol %, the flame propagates with axial symmetry after a delay. II. In the range 5.5-5.2 vol % of oxygen, a reaction starts near the wire after about the same delay as in I, followed much later by ignition below the wire and a flame which also spreads upwards. III. Below 5.2 vol %, a reaction starts after about the same delay as in I but instead of a propagating flame, a reaction zone is stabilized near the wire. The delays and to some extent the ranges of composition are dependent on the temperature of the wire. In Fig 5, the delays and the burning velocities derived from measurements of flame velocities on the Schlieren pictures are shown. By determining the optical path length of a point on the interferograms away from the temperature field around the wire as a function of time, the density and hence, by considering an adiabatic compression, the pressure and the temperature of the unburned gas can be determined. By restricting the measurements to within 2 cm of the wire, the pressure can be regarded as being constant and a simple expression $S_u = S_s \rho_b / \rho_u$ [7] used to derive burning velocities from the observed flame velocity S_s , the density of the unburned gas ρ_u and the mean density ρ_b of the burned gas. The value of ρ_b is however difficult to determine [7] giving an uncertainty in S_u of about 30 %.

During the preignition period, interferograms observed with reactive gas mixtures are identical with those taken in pure hydrogen [3] as shown in Fig 3, indicating that during this period no significant reaction occurs. Due to the buoyancy induced, the outer isotherms immediately start to accelerate with a constant acceleration as

predicted in Ref 3. The convection currents around the wire during the preignition period can be reduced by accelerating the wire with an acceleration which can be predicted. As shown in Fig 4, an acceleration of the wire having no significant influence in range I, increases the ignition delay in range II.

Discussion

Experimental apparatus of different kinds has been used to measure the limits of flammability, often producing conflicting results[8,9]. A closed vessel, chosen in these experiments, seems to be preferred to tubes [9,10]. In a closed vessel the limits of flammability have been derived by certain criteria from detectable pressure increase [4] or conversion degree as a function of the composition of the mixture[3]. The finding of these experiments are:

- I) The convection currents have time to develop when the limit of flammability is approached and hence a comparison with thermal ignition theory (as done by Ashman and Büchler[11]) for stoichiometric hydrogen-air mixtures is not possible.
- II) After the limit has been passed convection currents feed unburned gases to the reaction zone near the wire. This results in a slow consumption of the content of oxygen and hence flammability limits should not be derived by conversion degree only as a function of mixture consumption.
- III) The motion of the flame kernel is komplex due to the induced convection currents and cannot be explained by the model proposed for spherical flames by Ref 6.

Acknowledgments

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Figure captions

Fig 1. Experimental apparatus.

Fig 2. Schlieren pictures and interferograms

a) $H_2+5.45$ vol % of O_2	$T_{\text{wire}} = 1325K$	scale 1:3
b) $H_2+5.05$ vol % of O_2	$T_{\text{wire}} = 1325K$	scale 1:3
c) $H_2+5.65$ vol % of O_2	$T_{\text{wire}} = 1425K$	scale 1:2
d) $H_2+5.35$ vol % of O_2	$T_{\text{wire}} = 1425K$	scale 1:2

Fig 3. Interferogram

a) N_2	$T_{\text{wire}} = 1166K$	
b) N_2	$T_{\text{wire}} = 1166K$	acceleration = 0.66 ms^{-2}
c) H_2	$T_{\text{wire}} = 1393K$	
d) $H_2+5.35$ vol % of O_2	$T_{\text{wire}} = 1425K$	acceleration = 0.71 ms^{-2}

Fig 4. Distance from the wire to the outermost fringe below as a function of time.

$T_{\text{wire}} = 1425K$

1) $H_2+5.7$ vol % of O_2	acceleration = 0.90 ms^{-2}
2) $H_2+5.65$ vol % of O_2	
3) $H_2+5.5$ vol % of O_2	
4) $H_2+5.45$ vol % of O_2	
5) $H_2+5.35$ vol % of O_2	
6) $H_2+5.35$ vol % of O_2	acceleration = 0.71 ms^{-2}

Fig 5. Delay time to the start of the reaction near the wire, convection currents at the start of the reaction 3 and laminar burning velocities as a function of the oxygen content.

Δ Jahr, given in Ref 1 . $T_{\text{wire}} = 1325K$, length of the wire = 36 mm.

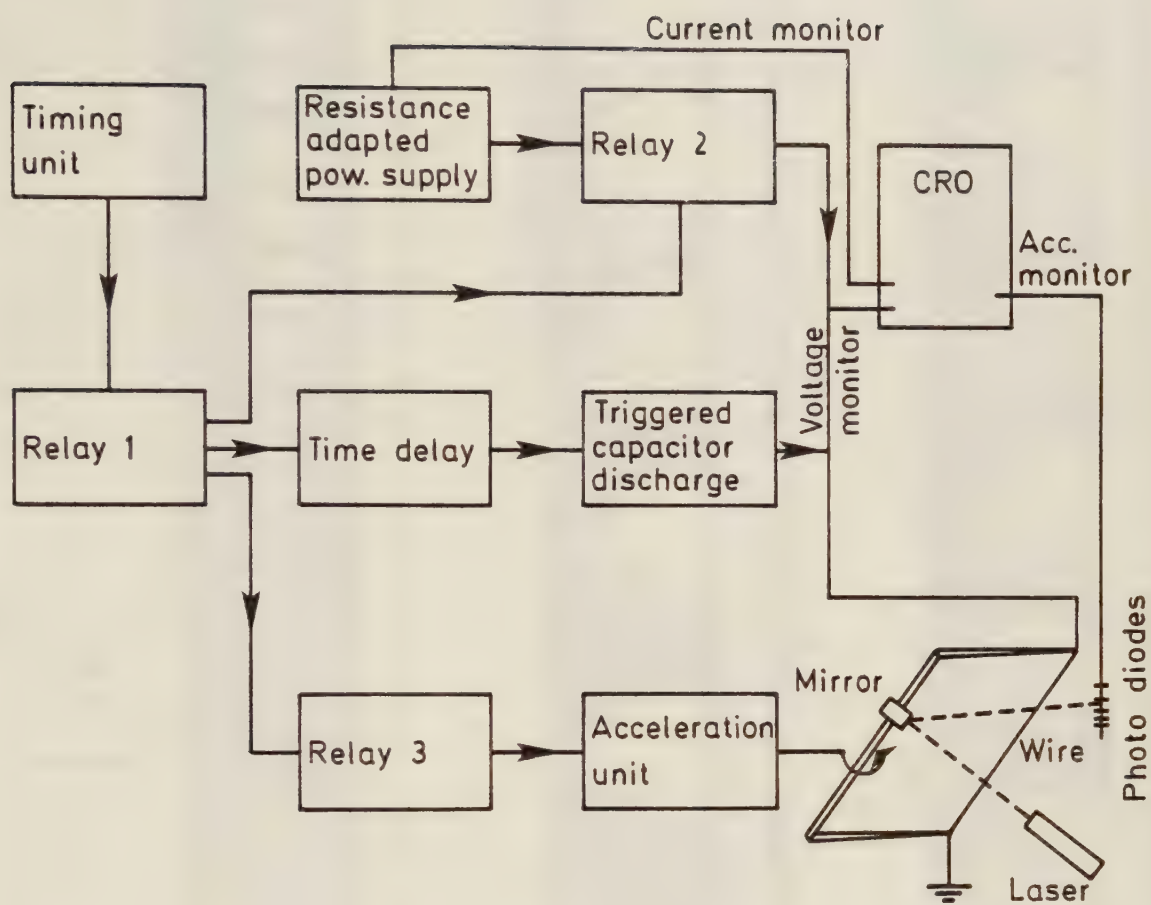


Fig 1

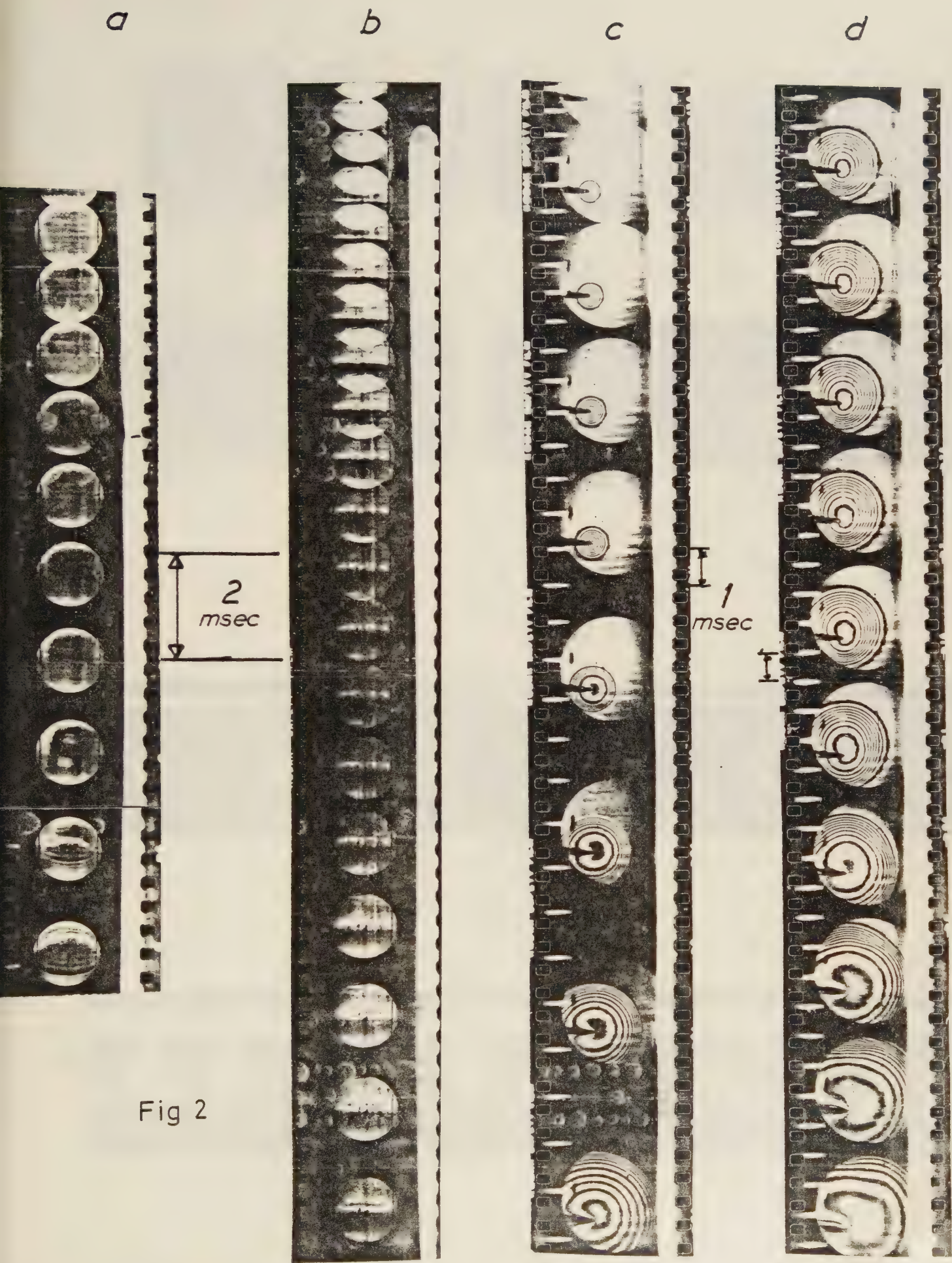


Fig 2

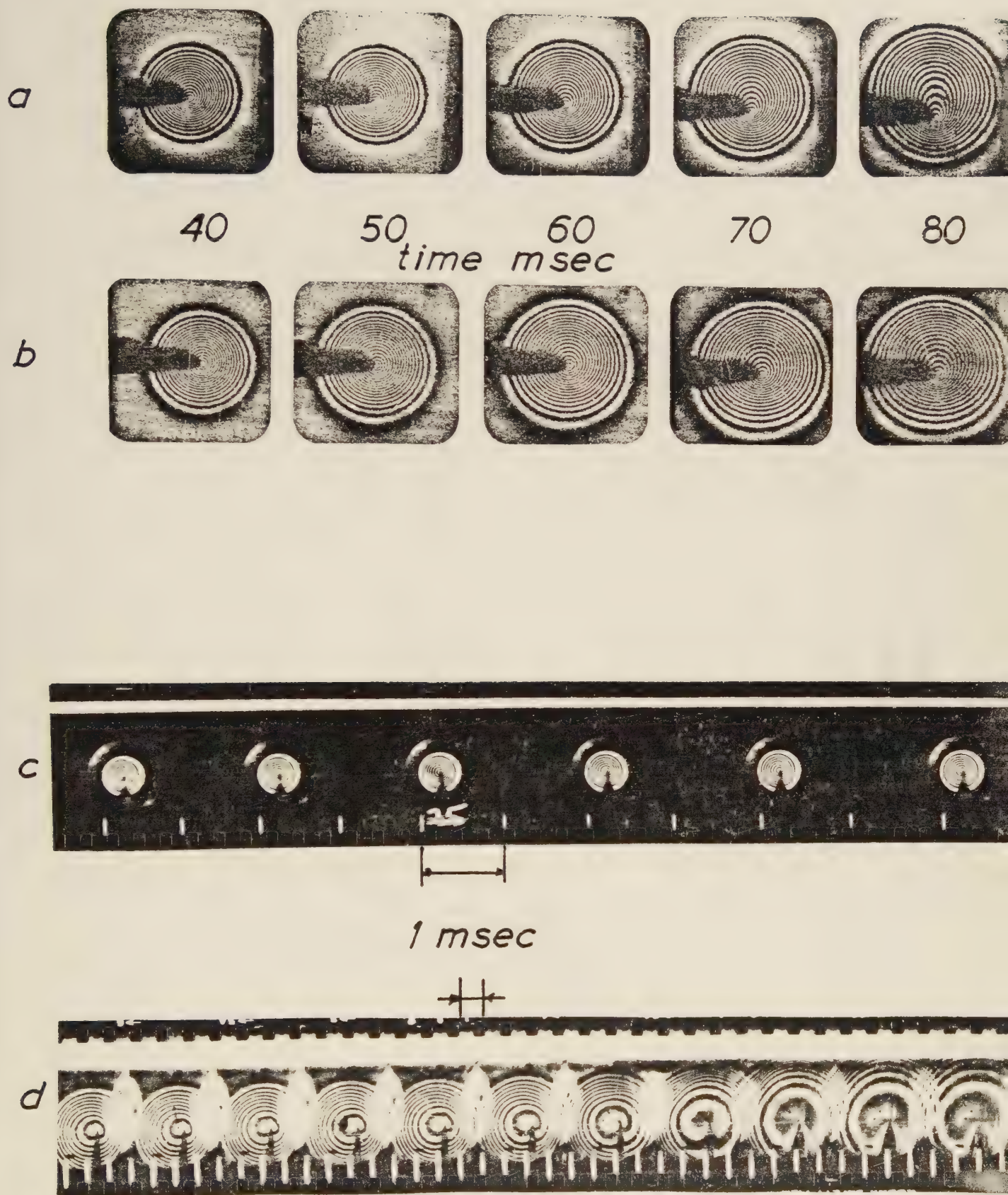


Fig 3

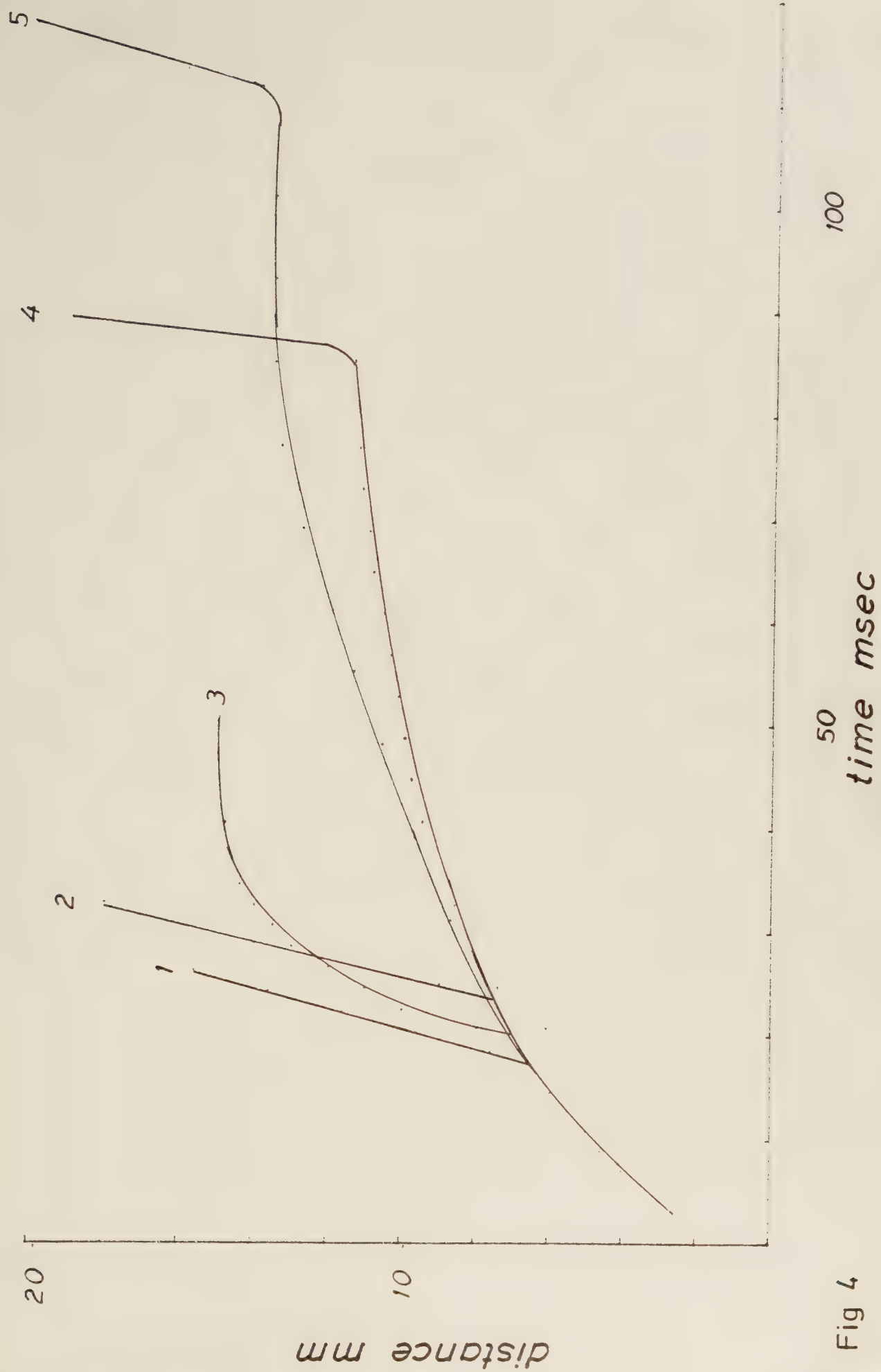


Fig 4

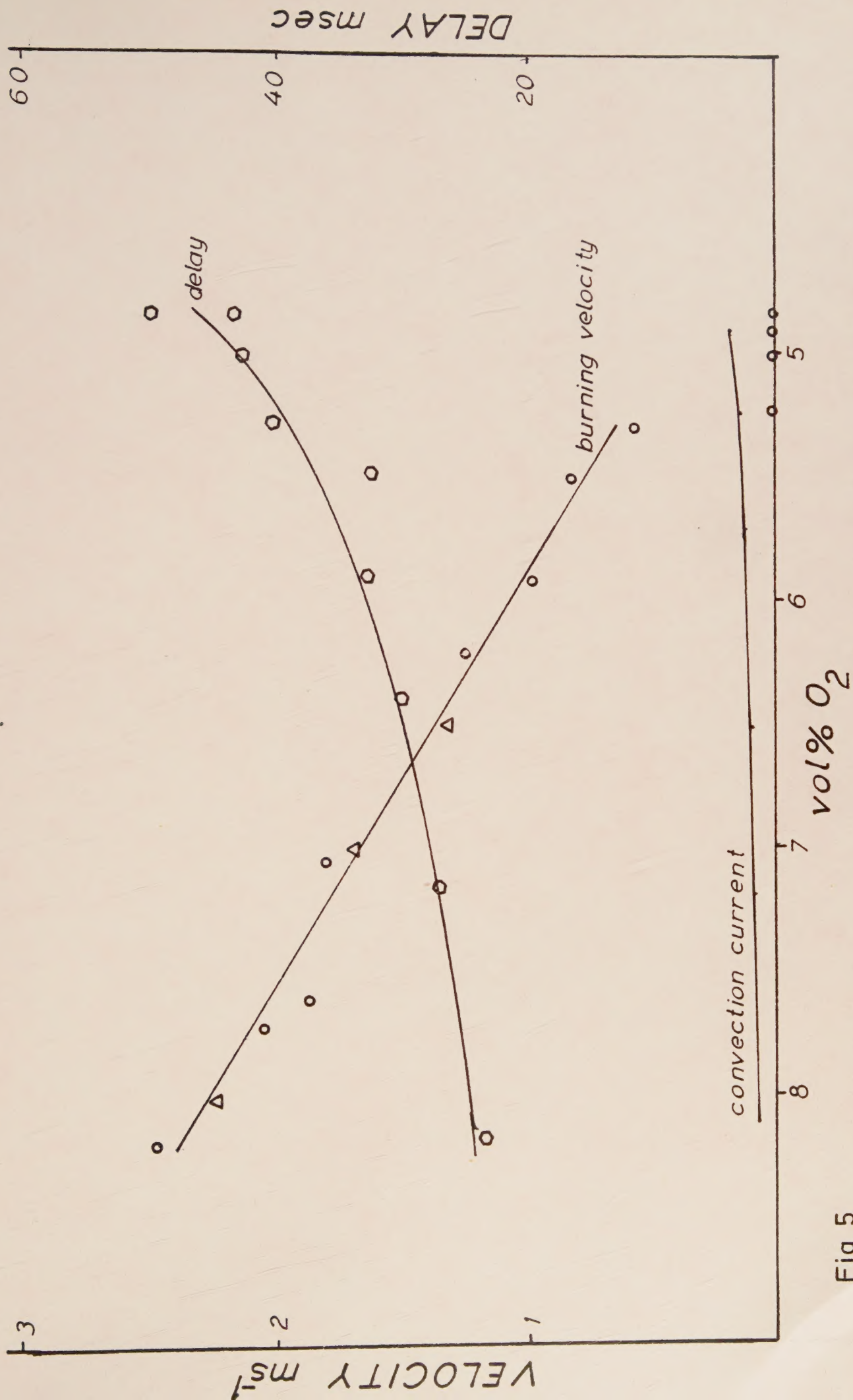


Fig 5

